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HYDROGEN GENERATION FOR FUEL CELLS

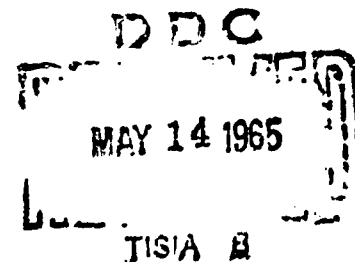
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REPORT NO. 8

CONTRACT NO. DA-36-039-SC-89077
TASK NO. 1G6 22001 A 053-04

FINAL REPORT
FOR THE PERIOD

1 MAY 1962 - 30 SEPTEMBER 1964



U.S. ARMY ELECTRONICS LABORATORIES
FORT MONMOUTH, NEW JERSEY

19991022181

ENGELHARD

I N D U S T R I E S, I N C.

PROCESS EQUIPMENT DIVISION
NEWARK, NEW JERSEY

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HYDROGEN GENERATION

FOR FUEL CELLS

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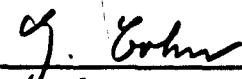
U.S. ARMY ELECTRONICS LABORATORIES

Object: To conduct investigations on the conversion of economical fuels to hydrogen for use as a fuel in primary fuel cell batteries.

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I. PURPOSE

The work undertaken in this contract consists of investigations to develop equipment for generating hydrogen from primary fuels in such a manner that the generation of the hydrogen and its separation from other reaction products can be accomplished easily in the field on a portable basis.

An experimental generator is to be designed, fabricated and delivered to U.S. Army Electronics Laboratory with the necessary reactants to produce 0.25 pounds of hydrogen distributed over a 12 hour period. The device, including fuel for 12 hours operation, is not to exceed 15 pounds in weight or a 2-cubic foot volume.

The experimental generator must be easily adaptable to integration with a 200 watt, 28 volt hydrogen-oxygen fuel cell battery.

Ammonia and hydrocarbons are to be investigated with an attempt to secure fuel cell operation at practical fuel cost.

The development of an experimental liquid hydrocarbon fueled hydrogen generator is to be undertaken with the goal of a lightweight portable unit capable of producing 10 SCFH of hydrogen for use with a 500 watt, hydrogen-oxygen, fuel cell. A breadboard hydrogen generator assembly is to be fabricated and delivered which represents the work accomplished under this contract.

II. ABSTRACT

Investigations were undertaken to develop small portable hydrogen generators for fuel cell use. Fuel sources considered included ammonia, methanol, and liquid hydrocarbons. The Engelhard Hydrogen Process combines three basic concepts: a hydrogen producing reaction, the removal of the pure product hydrogen, and the utilization of the residue off gas as fuel to supply the energy requirements of the process. The hydrogen product is withdrawn by means of diffusion through a palladium alloy and is, therefore, of highest purity.

Initial investigations were performed with ammonia as a fuel to establish the feasibility of the process and to establish design parameters for prototype generators. Several ammonia fueled prototype generators were constructed, tested, and modified to reflect design improvements. An ammonia feed system was developed which uses the heat content of the waste stack gas to vaporize the liquid ammonia feed.

A final prototype portable ammonia fueled hydrogen generator was built and delivered to the U. S. Army Electronics Laboratory. This self-sustaining unit produced .27 lbs. of hydrogen over a 12-hour period from approximately 2.5 lbs. of ammonia with a corresponding thermal efficiency of over 70%. This generator met the contractual requirements except for slight excess weight. It could, however, be produced substantially lighter by using specially fabricated components instead of standard commercial items.

Life studies were performed with ammonia fueled generators. One operated for 490 hours before mechanical failure and another completed 87 cycles for 1035 hours of operation at full rated capacity before failure due to corrosion at the inlet of the reactor tube.

Investigations have shown that it is possible to steam reform methanol at an equimolar water ratio and obtain conversion levels and hydrogen yields sufficient to operate a generator on a self-sustaining basis.

Due to their universal availability, hydrocarbons are desirable fuels for hydrogen generation. These are the only fuels which would meet the goal of a fuel cost of less than 20 mils per KWH. Experimental work was performed to show that liquid hydrocarbon fuels could be steam reformed to produce hydrogen at thermal efficiencies such that portable, lightweight, self-sustaining apparatus could be developed.

Catalyst life tests were performed which established that liquid hydrocarbons such as BTX UDEX Raffinates or JP-4 Jet Fuel can be steam reformed at low water to carbon ratios with reasonable catalyst life, provided the sulphur content is below certain specified limits.

Detailed design studies have been performed on liquid hydrocarbon consuming units at the 4 SCFH level (200 watt) and at the 10 SCFH level (500 watt). Results of the studies demonstrated reasonable sizes and weights.

Components for a hydrocarbon fueled generator were investigated to establish their availability and reliability.

Two experimental liquid hydrocarbon fueled hydrogen generators have been fabricated and tested. Hydrogen production rates of 10 to 14.5 SCFH were realized with gross heating value efficiencies of 52 to 71%. Start-up was accomplished within 30 minutes.

One generator was equipped with an integral air pressurized feed system and mounted as a breadboard assembly on a panel. The unit was delivered to the U. S. Army Electronics Laboratory, where a demonstration run was performed. Within the contract period it was not possible to eliminate certain instabilities in the system. Additional engineering would be required to construct a field unit.

A primary fuel cost of 17 mils per KWH has been established using liquid hydrocarbon fuel.

III. PUBLICATIONS, LECTURES, REPORTS AND CONFERENCES

Publications and Lectures

On May 19, 1964 a paper "Hydrogen Generators", describing work on this contract was presented by Mr. L. E. Goodman at the session on fuel cell batteries at the Eighteenth Annual Power Sources Conference at Atlantic City, New Jersey.

Reports

Quarterly Reports Nos. 1 through 7 on "Hydrogen Generation for Fuel Cells," contract No. DA-36-039 SC-89077.

Conferences

A conference was held on March 5, 1964 at the U. S. Army Electronics Laboratory, Fort Monmouth, New Jersey, attended by Mr. A. F. Daniel, Dr. H. F. Hunger, Mr. S. Bartosh and technical personnel of the contractor to discuss the status of present efforts, program for the next quarter, and extension of the contract to 30 June, 1964.

A conference was held on June 23, 1964 at Engelhard Industries, Inc., attended by Mr. S. Bartosh and technical personnel of the contractor to discuss the status of work done and extension of the contract to 30 September 1964.

A conference was held on September 24, 1964 at Engelhard Industries, Inc., attended by Mr. S. Bartosh and technical personnel of the contractor to discuss the status of the contract and delivery of the Breadboard Hydrogen Generator Assembly.

On October 20, 1964 the Breadboard Hydrogen Generator PT-9 was delivered to the U. S. Army Electronics Laboratory, Fort Monmouth, New Jersey. The unit was operated for Mr. Bartosh, Dr. H. F. Hunger, Mr. A. F. Daniel, and others.

IV. FACTUAL DATA

A. Operating Principles

Investigations were undertaken to develop small portable hydrogen generators for fuel cell use. Fuel sources considered for the generation of hydrogen included ammonia, methanol, and liquid hydrocarbons. In the case of ammonia the hydrogen production is by catalytic thermal dissociation, and with methanol and hydrocarbons by steam reforming.

In either case, the Engelhard Hydrogen Process is used in which the pure product hydrogen is drawn off through a palladium alloy diffuser, and the remaining off gas is utilized to provide the heat for the generating process.

In addition, because only hydrogen passes through the diffusion element, no further purification equipment or processing is required. The resulting equipment, therefore, can be extremely compact as well as highly efficient, and lends itself well to miniature equipment.

B. Ammonia as a Fuel for Hydrogen Generation

Ammonia when dissociated, produces a gas mixture containing 75 per cent hydrogen. Part of the hydrogen produced is separated by diffusion and the residue off gas burned to provide the necessary heat to maintain the process. Our design for a unit of 4 SCFH capacity presently provides for separating approximately 50 per cent of the hydrogen available and to use the balance for producing the heat to maintain the process. A catalyst is used to burn ammonia in air to nitrogen and water during start-up.

There are, therefore, two separate catalyst chambers in the generator; first, a combustion chamber to provide the heat necessary for dissociation, and second, a reaction chamber to split the ammonia. The separation of the hydrogen from the hydrogen-nitrogen mixture is accomplished by a palladium alloy diffuser which is suitably arranged for exposure to the hot reaction gases.

1. Process and Component Studies

In conformance with the technical guide lines of PR&C No. 62-ELP-D-4208 requiring the production of 0.25 pounds of hydrogen over a 12 hour period, the following limits were specified.

$$\text{H}_2 \text{ Produced, SCFH} * = \frac{0.25 \times 359 \times 520/492}{2 \times 12} = 3.96$$

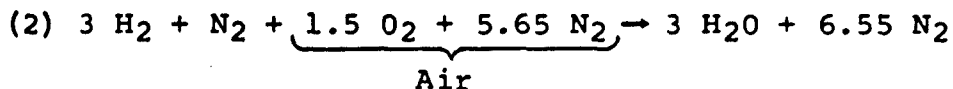
Lbs. NH_3 /hr: 0.24 (2.88 Lb/12 hr. period)

NH_3 Pressure - 35 psig (Minimum ambient temperature of 22°F)

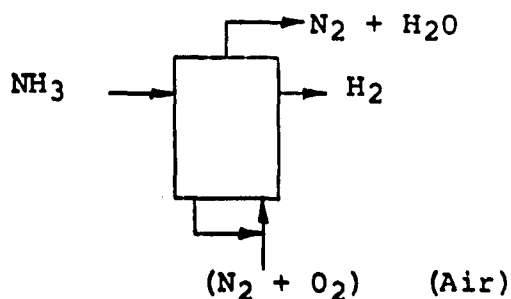
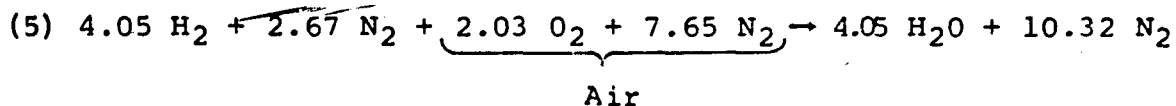
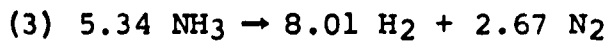
Total Volume - 2 cubic feet

Total Weight - 15 pounds

The basic reactions in the system are:



Total reactions for the system are:



* Standard Conditions established at 60°F and 29.92" Mercury.

For the development of the generator, several components were studied separately prior to assembly of a complete generator. Based on these results, materials were selected, parts designed and the first prototype generator was constructed.

a. Aspirator

Ammonia is supplied to the unit at elevated pressure. After dissociation and withdrawal of product hydrogen the pressure of the residual gas is utilized to aspirate air required for the combustion.

During start-up the fuel gas flowing through the orifice into the combustion chamber is ammonia. As the unit warms up and comes into operation the fuel gas composition gradually changes to a mixture of 60 percent hydrogen and 40 percent nitrogen. At the outset, the fuel gas is delivered at ambient temperatures but it is gradually heated up. At steady state conditions, a delivery temperature of 1100°F was expected.

A laboratory investigation of aspirators was performed using both ammonia and a synthetic off gas. design parameters included aspirator throat diameter and orifice diameter. The test apparatus is shown in Figure 1 and results obtained with the synthetic off gas are shown in Figure 2.

b. Combustion Chamber

The test combustion chamber, Figure 1, was operated using both ammonia and synthetic off gas as the fuel and temperature profiles were obtained.

c. Igniter

When starting the generator with ammonia at ambient temperature, a localized hot spot has to be provided temporarily. Catalytic combustion

of ammonia to nitrogen and water requires heating to approximately 1200°F. Using the test combustion chamber, successful start-ups were obtained using a coil of 4 mil diameter wire of 90% platinum--10% ruthenium alloy as an igniter as shown in Figure 3.

The original igniter system did not prove suitable when subjected to repeated testing. The coil of the igniter was too fragile for this application. In addition, the catalyst pellet sintered under operating conditions. To select a suitable igniter system, various combinations of wire were tested by measuring the impressed voltage and current drain for the first visual sign of glowing across a coil made from a one-inch length of wire. Various batteries were also tested for their applicability. The best results were obtained with a bare coil of 10 mil diameter wire, 90% platinum--10% rhodium with pure silver lead wires using two 1.5 volt manganese dioxide alkaline C batteries connected in series. This system is shown in Figure 4.

Only a 4-second ignition period was required to start the combustion of ammonia and better than 10 start-ups could be obtained from a battery.

An auxiliary igniter was developed for use in the event of malfunction of the coil igniter system or batteries. This consists simply of a 1/16" diameter metal rod preheated by a single match or by the flame of a cigarette lighter for about 15-20 seconds. The lighter is probably preferable since it is more reliable under a variety of weather conditions. Easy access to the combustion tube is achieved through a Swagelok fitting. The ammonia feed is turned on and the pre-heated metal rod inserted into the combustion tube through the fitting and held in intimate contact with the catalyst for about 10 seconds. After ignition is secured, the Swagelok fitting is re-tightened. Repeated start-ups have been accomplished utilizing this method.

d. Ammonia Feed Regulator

Testing of the ammonia fueled hydrogen generator systems revealed inadequate sealing within the ammonia feed regulator. The manufacturer was contacted and the metal seat was replaced with a ~~soft~~ (Buna-N) seat. This modified Fisher Governor Regulator, Model 67-9 then proved to be satisfactory.

2. First Prototype Hydrogen Generator

A test reactor was set up as shown in Figure 5. The weight of the first generator without insulation housing or accessories was 2.6 pounds. A commercial ammonia cylinder was used instead of a portable container. In the initial tests, the prototype hydrogen generator produced pure hydrogen at a rate of 0.32 pounds/12 hours, used ammonia at a rate of 3.8 pounds/12 hours and had a fuel off gas analysis of 61.3% hydrogen (design 60%). Starting at room temperature, ignition was secured within 35 seconds, hydrogen flow started in 7 minutes and was at full flow rate after approximately 35 minutes.

3. Second Prototype Hydrogen Generator

A modified hydrogen generator was built using readily available standard materials which were unnecessarily thick. The resultant weight was approximately 6.5 pounds.

Since cyclic operation would produce more severe stresses in the reactor walls it was preferable to continuous operation at this stage of development. The reactor was started up every morning and shut down every evening.

A summary of the test data for this hydrogen generator is shown in Table 1. Hydrogen was produced at an essentially constant rate of 0.28 pounds/12 hours calculated off gas composition ranged from 54.6 to 59.5 percent hydrogen (design maximum 60%).

The combustion tube failed after Run No. 5. An examination of the tube indicated a circumferential fracture in the area of the flame. This was caused by thermal expansion of the material which effected compression forces during heating, resulting in some plastic creep. The tube then went into tension on cooling and this sequence was repeated with each operating cycle. The reactor was rebuilt with the addition of a heat distributor, which consisted of a heavy metal annular core separating the flame from direct contact with the combustion tube. The heat distributor reduced the maximum temperature of the hot spot of the combustion tube at the flame, and distributed the available heat over a longer length. During Runs No. 6 through 11, the reactor was operated with the heat distributor in the combustion chamber.

In early tests, the hydrogen generator reached full flow rated capacity within 30 minutes without a heat distributor and somewhat longer with a heat distributor.

4. Generator Performance Data

Test reactor PT-3 was operated to secure life and performance data. The apparatus was arranged as shown in Figure 6. Operation was on a daily basis, 8 hour on-16 hour off cycle, with complete cooling in the off period. The generator failed during the 21st cycle due to thermal expansion stresses.

During the period of testing PT-3, a modified test generator for producing hydrogen from ammonia designated PT-5 was designed and built. This generator was designed without a heat distributor such as used in PT-3. Instead, it utilized a heavier walled combustion section to distribute the heat.

In initial tests of PT-5, the generator developed a leak, during the second cycle, between the reactor and the pure product hydrogen chamber. The leak was found to be at the weld of a flange to the combustion chamber. Since the defect developed at the junction between two components and not in a component itself,

it was concluded that the remaining problem was one of assembly techniques. The generator was modified and reassembled, with the feed gas flow now counter-current to the stack gas flow. The generator was operated again to check start-up times and the reactor performance.

Typical test data are shown in Table 2. Typical temperature profiles for PT-3, PT-5 parallel flow, and PT-5 countercurrent flow are shown in Figures 7, 8 and 9 respectively. The temperature distribution curves shown on Figures 7, 8 and 9 show that all points of the systems studied had temperatures within the range required for proper operation of the generator.

A generator, designated PT-3A, was constructed in which were combined the improved mechanical features of PT-5 and the attractive operating characteristics of PT-3. This unit was run for a total of 11 cycles and an accumulated operating time of 169 hours. At this time there was a weld failure in the diffusion section. The unit then was modified successively into reactors designated PT-3B, PT-3C, PT-3D. In each of these modifications the reactor configuration was changed with regard to the location of the diffusion section, preheating and location of the ammonia feed line, size and location of the cracking chamber and the direction of the ammonia feed flow in the catalyst reaction section. Typical test data for these units are shown in Tables 3 and 4.

On the basis of the test results, test reactor PT-6 was built with wall thicknesses changed to reduce the thermal stresses set up during operation. The ammonia flow is admitted at the bottom of the reaction zone at the hottest part of the combustion chamber in order to control the maximum temperature with the cool ammonia feed stock. The location of the ammonia feed at this point provides for rapid heating of the incoming ammonia and distributing the temperature more evenly throughout the reaction zone. Tests on this unit PT-6 indicated satisfactory performance as regards the quantity of hydrogen produced. Typical test data is shown in the Test Data for Ammonia Fuel,

Table 5. However, the start-up time was too long, probably due to the increased mass of the material in the generator.

5. Ammonia Feed System

The ammonia feed system must provide vapor at a sufficient pressure for operation at various ambient temperatures. In order to keep the weight of the ammonia carried for 12 hours operation to a minimum, it is desirable to have as little liquid as possible remaining in the container at the completion of the run. Initial tests were performed with a small ammonia container holding the fuel supply for 12 hours operation of the generator. Within 2 hours the pressure had dropped from 125 psig to 35 psig indicating insufficient transfer of heat from the ambient room-temperature air to the ammonia liquid. The limiting heat transfer area is the surface of the ammonia container wetted with liquid ammonia.

The energy requirements for the vaporization of the liquid ammonia were calculated to be 105 to 120 BTU per hour, and the available heat content of the waste stack gas was calculated to be approximately 240 BTU per hour. It was decided, therefore, to investigate the feasibility of a heat exchange device which would employ the heat content of the waste stack gas to vaporize the ammonia feed.

a. Ammonia Vaporizer

During several tests on PT-5, the equipment was arranged as shown in Figure 10. The stack gas was conducted through a coil which surrounded a small ammonia container, five-inch diameter by seven inches long. The container and coil were lightly insulated. This established the feasibility of using the waste stack gas to vaporize the liquid ammonia.

b. Ammonia Container

A Standard Military oxygen tank Type A-6 was modified and fitted with an outer shell of two

spun aluminum heads which were welded together as shown in Figure 11.

In operation, the waste stack gas was admitted to the upper side tube and the gas passed around the container and down through the space between the container and jacket. The liquid was heated in this manner to provide the heat of vaporization. Any water condensed dripped out of the bottom port with the vented stack gas.

It was found that in order to maintain the pressure in the container below 250 psig it was necessary to install a vent in the stack gas transfer line and to by-pass a portion of the stack gas.

c. Ammonia Separator

A teflon coated ceramic candle with a 1.4 micron porosity was evaluated for use as a means of separating ammonia liquid from ammonia vapor so that the feed tank could be operated in other than the vertical position. It was found, however, that in any position where the liquid ammonia contacted the candle separation did not take place and the device was unsatisfactory.

6. Sample Hydrogen Generation Device

Based on a detailed review of the data, reactors PT-7 and PT-7A were constructed for delivery of one unit to the U.S. Army Electronics Laboratory. These two units were built using lighter materials of construction and incorporated a bellows assembly to allow for thermal expansion of the chambers. The assembly configuration of the complete hydrogen generator is shown in Figure 12. A component weight summary is provided in Table 6. Significant further weight savings could be achieved by using specially fabricated parts instead of the standard commercial components used in the prototype.

a. Performance Characteristics of Delivered Generator

Hydrogen generator PT-7A was tested in our laboratory prior to delivery to the U.S. Army

Electronics Laboratory. It produced pure hydrogen at a rate of 0.272 pounds per 12 hours, used ammonia at a rate of 2.5 pounds per 12 hours and had thermal efficiencies of 73%. The thermal efficiency is defined as the ratio of the gross heating value of the product hydrogen to the gross heating value of the ammonia feed. Starting up at room temperature, ignition is secured in 4 seconds; one-half capacity is reached in 15 minutes; and full rated capacity, 4 SCFH, in 30 minutes. Within 50 to 60 minutes, the ammonia start up pressure of 35 psig is reduced to 23 psig to maintain the pure product hydrogen rate at 4 SCFH. This is done to optimize performance of the unit by conserving fuel and operating at higher thermal efficiencies. This generator, therefore, has the ability to yield a relatively broad range of product hydrogen flow rates dependent on the operating pressure. Typical performance data are shown in Table 7.

7. Ammonia Fueled Generator Life Study

Tests were performed to obtain a life study and to improve and modify procedures and ancillary equipment. The apparatus was arranged as shown in Figure 13. The operation was on a daily basis, approximately 12-14 hours on, 12-10 hours off cycle, with complete cooling in the off period. In some tests ammonia was drawn directly as vapor from a 150 pound cylinder. In other cases, the small portable container shown in Figure 11 was used and the stack gas was utilized as a heat source for vaporizing the liquid ammonia.

a. Portable Hydrogen Generator, PT-7

This generator was similar to the prototype PT-7A delivered to the U.S. Army Electronics Laboratory, Fort Monmouth. Typical test data for this hydrogen generator device are shown in Tables 8 and 9. The generator had the ability to deliver more than 4 SCFH of pure hydrogen over a 12 hour period from about 3.0

pounds of ammonia. It should be noted from the data that the thermal and material efficiencies for this device, PT-7 are somewhat lower than those previously reported for the sample generator, PT-7A delivered to Fort Monmouth. This was due to the fact that the generator had less diffusion surface than PT-7A. It was, therefore, necessary to operate with a correspondingly greater ammonia feed rate than generator PT-7A.

This hydrogen generator also has the ability to deliver a relatively wide range of product hydrogen flow rates dependent on the operating pressure. This varies from about 5.5 SCFH at 35 psig to about 4.2 SCFH at 26 psig and lower flow rates at lower regulator settings.

~~The test~~ generator was run for a total of 39 cycles with an accumulated operating time of about 490 hours. There was no indication of any deleterious effects due to this cyclic operation. However, during the start-up of the 40th cycle it was noted that a leak had developed into the pure hydrogen chamber. Examination of the unit after dissection revealed that the failure had occurred in a welded joint of the diffusion membrane to the header.

b. Portable Hydrogen Generator, PT-6

Instead of repairing PT-7 life studies were continued with reactor PT-6. This generator is nearly identical in configuration to PT-7. The operation was again performed on a cyclic basis. The performance of this generator obtained during the first 20 cycles of operation has been reported in Table 5.

The generator was operated for 87 cycles with an accumulated operating time of 1035 hours. Up to this point there was no indication of any deleterious effects. The generator performance continued to meet design capacity specifications of greater than 4 SCFH of hydrogen as shown in

typical test data listed in Table 10 and 11.

This device also was capable of delivering a relatively broad range of product hydrogen rates dependent on the control pressure settings. Its material and thermal efficiencies were somewhat lower than those for PT-7. This is due to greater heat losses associated with the thicker materials of construction of PT-6.

It should be noted that the generator had been X-ray examined after 20 cycles and again after 40 cycles. There was no evidence of any unexpected distortions noted in the reactor system either time.

On the start-up of cycle No. 88, a leak of ammonia into the pure hydrogen side of the generator was found causing termination of the testing.

The unit was dismantled and X-ray examined; no evidence of excessive distortion was found; nor was any point of failure observed. The reactor was leak tested, using nitrogen at pressure, and a leak was found in the wall of the reaction chamber at the point where impingement of the ammonia feed occurred. The reactor tube is of type 316 stainless steel and the surface area in the vicinity of the leak was porous and pitted.

The remaining parts of the generator, including the palladium alloy diffuser, were tested and were found to be free of leaks.

Sections were taken for metallographic examinations of the reactor tube at the area of the failure near the bottom, Figure 14 and at the upper portion of the tube away from the area of failure, Figure 15.

Figure 14 shows the nitrided area of failure. The metal is covered with a friable dark layer which appears very dark gray (Zone A).

Immediately under the dark phase is a very hard non etching white region which does not show grain boundaries (Zone B). The remaining "good" metal is shown as Zone C. In Figure 15, no evidence of nitride formation is found. This would indicate that failure was due to the formation of nitrides by the pure ammonia feed impinging on the reactor wall, probably in conjunction with thermal stresses present at this area. (A bellows assembly was used in subsequent designs to eliminate the thermal stresses).

Although type 316 stainless steel is reported to be suitable for use in ammonia converters a check of the literature after this failure showed that recent corrosion tests of the 300 series stainless steels in pure ammonia pipeline service with turbulent flow at about 1000°F indicated type 316 had an excessive rate of formation of nitrides.¹ The nitriding tendency of an alloy should increase with the presence of additional nitride-forming elements in a given base (such as molybdenum in type 316 stainless steel). It is suggested that the molybdenum may contribute to the development of an unusually friable or exfoliating scale which could spall excessively under the action of a turbulent gas stream.²

It is recommended that a higher nickel alloy (such as inconel) be used for resistance to nitride formation by pure ammonia at high temperature.³

1. "CORROSION RESISTANCE OF THE AUSTENITIC CHROMIUM-NICKEL STAINLESS STEELS IN HIGH TEMPERATURE ENVIRONMENTS" The International Nickel Company, Inc., New York 5, New York 1963
2. "BEHAVIOR OF STAINLESS STEELS AND OTHER ENGINEERING ALLOYS IN HOT AMMONIA ATMOSPHERES", CORROSION, Vol. 17, No. 4, pp. 191t-195t (1961) April
3. Personal Communication, Huntington Alloy Div., International Nickel Company, Inc.

C. Methanol as a Fuel for Hydrogen Generation

The direct dissociation of methanol to carbon monoxide and hydrogen would be inefficient. After removal of some hydrogen by diffusion the residual carbon monoxide-hydrogen mixture, when used as a fuel gas, will contain more energy than is required for operating the process. For this reason, reforming of a concentrated solution of methanol and water offers advantages. This will provide additional hydrogen and the remaining mixture, after diffusion of hydrogen, has a heat of combustion in balance with the heat requirements of the system.

Steam reforming of methanol, as practiced in the past; required the use of a large excess of water so that the water-gas shift reaction would be essentially complete. It was desirable to convert the carbon to carbon dioxide as completely as possible because removal of carbon monoxide from the net product hydrogen was not feasible. In the Engelhard process, a large excess of water is not required because it can tolerate moderate amounts of carbon monoxide. Further, hydrogen can be separated by diffusion as easily from a carbon monoxide mixture as a carbon dioxide mixture. In addition, the energy content of the carbon monoxide is not lost; it can be used as fuel in the combustion zone to operate the process.

One possible composition is the equimolar mixture of water and methanol. Our laboratory test work was aimed at verifying that this composition could be successfully reformed to yield a stream capable of being separated into pure hydrogen and a balanced fuel gas supply.

1. Experimental Apparatus

The test reactor, vaporizer and associated equipment were arranged as shown in the Schematic Flow Diagram, Figure 16. A precious metal catalyst was used and the reaction heat was provided by external electrical heaters. In this test the hydrogen was not separated from the product gas since this test was conducted solely to obtain process design data for the reforming of methanol as a hydrogen source. The flow rate of the aqueous methanol solution was measured volumetrically. The volume of the gaseous products was measured by a

wet test meter and the steam in the effluent off-gas was condensed and measured by weighing. The effluent off-gas was analyzed using an Orsat apparatus.

2. Experimental Procedure

A single feed stream of an equimolar mixture of water and methanol (.444/1 volumetric ratio) was used. The system was purged with nitrogen while reaching the desired temperature and pressure conditions. During the run, conditions were held constant with the pressure at 150 psig and the temperature in the vaporizer and the reactor at 1000°F. The feed was brought into the system by means of the diaphragm pump, it was vaporized and fed into the reactor as vapor. The system pressure and temperatures of the vaporizer and the reactor were monitored and controlled manually.

The system was operated for 13 hours and shut down for an examination of the catalyst bed. The unit was then reassembled, and the test continued for an additional 55 1/2 hours until terminated by choice.

3. Experimental Test Results

The experimental test results obtained are listed in Table 12.

Based on the time of operation and the amount of feed stock, the catalyst loading was 79 grams of methanol per gram of catalyst. Constant performance was obtained throughout the test run. The product gas quantity varied between 39 and 45 SCFH for 1300 cc/hour of methanol-water mixture. Hydrogen was produced at a rate of 28 to 31 SCFH. No plugging occurred nor was there any evidence of coke formation.

The Orsat analysis data were incomplete, but a review of the data on the basis of equilibrium constants indicates that methanol first dissociates to carbon monoxide and hydrogen forming subsequently hydrogen and carbon dioxide by the water-gas shift reaction. The catalyst, while not establishing

equilibrium of the water gas shift does promote the reaction to a significant degree. Additionally, the catalyst used does not significantly promote formation of methane or hydrocarbons.

D. Hydrocarbons as Fuels for Hydrogen Generation

Hydrocarbons are desirable fuels for hydrogen generation due to their universal availability. They, however, present special problems in developing a portable, lightweight self-sustaining apparatus. Steam reforming requires the use of water which must be kept to a minimum. Fairly complex controls are necessary to assure stable and safe operation, all of which must be miniaturized.

Catalytic steam reforming of hydrocarbons consists of reacting the hydrocarbon fuel and water in suitable ratios in the presence of a reforming catalyst.

The reforming reaction produces hydrogen, carbon monoxide, carbon dioxide, and methane; in addition, the product gases contain excess water. In the Engelhard Hydrogen Process, the product hydrogen is withdrawn through a diffusion element and the remaining hot reactor gases are used directly as fuel, supplying the heat requirements for the process.

1. Exploratory Investigations

In order to obtain process information, exploratory experiments were performed. An externally heated reactor containing a commercial nickel catalyst was used and initial results showed that propane, No. 55 Naphtha, and JP-4 jet fuel could be steam reformed with high feed conversions. Typical results for JP-4 are shown in Table 13. However, using JP-4 as fuel, the life of the particular nickel catalyst employed was very short.

In a small portable field hydrogen generator, the injection of the liquid feed stocks should be as simple as possible. Consideration of the weight requirement for the equipment necessary to feed two

stream, either in the vapor or liquid phases, suggested the use of a prepared emulsion of liquid hydrocarbon and water as a single feed stream to minimize the required components. Experiments were carried out with emulsifiers for the JP-4/water system in suitable ratios. A summary of various emulsifier systems which have been screened is given in Table 14. These tests were performed with at least one part emulsifier per four parts of oil. The only agitation utilized was simple shaking by hand.

As the amount of emulsifier required was fairly large, and emulsions may not be stable below the freezing point of water, and as the addition of a third feed component (the emulsifier) would introduce additional logistic problems for a field unit, the investigation of emulsifiers was terminated.

2. Catalyst Life Testing

Life testing of reforming catalysts was performed with liquid hydrocarbon fuels in order to obtain process data.

a. Experimental Apparatus

The pumps, vaporizer, test reactor and associated equipment were arranged as shown in Schematic Flow Diagram, Figure 17. In these tests, the vaporizer and reactor were heated by external electric furnaces in order to provide close temperature control. The hydrogen was not separated from the product gas since the tests were conducted to obtain process data regarding catalyst life in reforming liquid hydrocarbon fuels. The flow rates of the fuel and water were measured volumetrically, and the volume of gaseous products was measured by a wet test meter. The product gas was analyzed using an Orsat apparatus and a gas chromatograph.

b. Experimental Procedure

The fuel and water were fed into the system, from separate burettes, by metering pumps. The

liquids were vaporized and fed into the reactor for reforming. The product gas was cooled first in an air cooled exchanger followed by an ice cooled condenser. The condensed water was withdrawn from the system. The product gas was sampled just ahead of the wet test meter. An ~~inert~~ gas loaded back-pressure regulator was used to maintain the system pressure.

c. BTX Udex Raffinate

BTX-Udex raffinate was chosen as the first hydrocarbon fuel for the testing. BTX-Udex raffinate is a selected naphtha, a by-product of petroleum derived benzene, toluene and xylene production and is in plentiful supply in refineries throughout the United States and Europe. Inspection data of the fuel used are given in Table 15.

The apparatus was operated at a 0.5 WHSV (gm oil/gm catalyst/hr.) at 1350°F and 150 psig. Using a molal water-to-carbon atom ratio of 3.0, the system was operated continuously for 480 hours with satisfactory results. The over-all composition of the product gas of this flow reaction corresponded to equilibrium temperatures somewhat lower than 1350°F. The deviation of the temperature is shown in Figure 18. The data shows that during continuous operation for 480 hours (with a loading of 250 g. oil/g. catalyst) the reactor was very close to equilibrium at all times.

After the run had been operated successfully on a continuous basis, a run was carried out involving daily cyclic operation, thus simulating the operation of a field unit.

For cyclic operation, the unit was cooled from operating conditions to 150°F and the pressure was released. The apparatus was then restarted.

This daily shutdown-start-up required approximately 4.5 hours with the unit operating at full rated capacity for 19.5 hours/day. Forty-eight stop-start cycles were performed during 1009 hours of actual operation. The data indicates that during this life test the reactor was close to equilibrium at all times.

d. JP-4 Jet Fuel

Although military specifications for jet fuels permit a sulfur content of 4000 PPM, at least one refining company (ESSO) processes their feeds in such a manner that based on their sales literature, a manufacturing specification of 400 PPM has been set. Several typical batches which we have purchased from this source have tested about 45 PPM of sulfur.

Further testing has shown that this grade of JP-4 can be steam reformed at the 45 PPM sulfur levels although higher temperatures are required. We, therefore, performed a continuous life test.

Inspection data of the fuel used are given in Table 16. The apparatus was operated at test conditions of a 0.5 WHSV (gm oil/gm catalyst/hr.), 1500°F, 150 psig, and a molal water to carbon atom ratio of 3.0.

After about 630 hours of normal operation, the water condensate which had been clear began to be cloudy. At about 850 hours the condensate began to show traces of oil which increased continually until the end of the run. The gas-make-rate, however, remained essentially unchanged throughout the run and the oil carrythrough which did occur would have remained a vapor and been burned in an operating unit. The gas analysis showed a slow increase in methane indicating deviation from equilibrium, and at about 950 hours a trace of ethane appeared in the off-gas indicating incomplete conversion.

Calculations of equilibrium temperatures are shown

in Table 17, in all cases the catalyst operating temperature was about 1500°F. Because the deviations of calculated equilibrium temperatures from operating temperatures were greater than the experimental errors after about 630 hours, it appears that the catalyst began losing activity at that time, which agrees with the qualitative observations mentioned above.

3. Design Studies

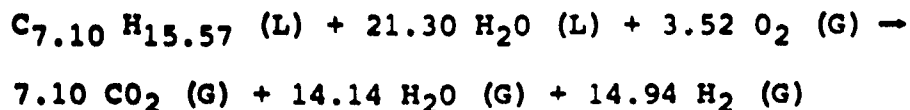
The purpose of the design studies was to design selected liquid hydrocarbon consuming units to the point where sufficient information was available to define the physical and operating characteristics, and to broadly specify component parts.

A heat and material balance study was conducted to determine optimum operating conditions for the process, the reactor-diffuser assembly was sized, and various materials of construction were investigated to obtain the smallest size and lightest weight configuration.

General design requirements may be calculated as follows:

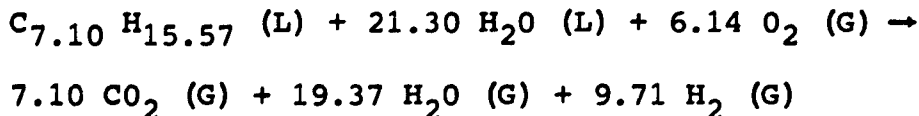
A typical BTX Udex Raffinate hydrocarbon fuel of 209°F average volumetric boiling point and 70.9° API gravity has an average molecular weight of 101, a hydrogen content of 15.5% and a liquid gross heating value of -21,000 BTU per pound. This permits calculation of an empirical formula of $C_{7.10} H_{15.57}$ and a heat of formation in the liquid phase at 77°F of -30.0 KCAL per gram mol.

The over-all reaction of steam reforming followed by the combustion of the non product reactor effluent may be written as follows:



This equation is based upon a 3.0 to 1 water to carbon molar ratio and an over-all heat of reaction of zero, which determine the maximum yield of hydrogen. This

theoretical yield cannot be obtained in actual equipment. Yields of 60 to 75% are considered feasible from an engineering standpoint. At 65% yield the equation becomes:



This over-all material balance is illustrated in Figure 19. This equation has a heat of reaction at 77°F of -544,000 BTU per pound mole of BTX Udex Raffinate fuel.

The incoming air, the fuel, and the water are assumed to be at the base temperature of 77°F. Based upon one pound mole of fuel, the heat of reaction then goes to:

- heat the product hydrogen to its release temperature of 1000°F before external cooling (63,000 BTU)
- heat the flue gas to its release temperature of 350°F (134,000 BTU, when using 50% excess air for combustion).

This leaves 347,000 BTU available for radiation and convection losses.

In the specific instance of a 10 SCFH generator which would supply a fuel cell of about 500 watt capacity this becomes 940 BTU per hour which is considered reasonable from an engineering standpoint. For this particular size generator these calculations give a requirement of 0.30 pounds of fuel and 1.10 pounds of water per hour.

Design studies have been prepared at the 4 SCFH level (200 watt) and at the 10 SCFH level (500 watt). The 4 SCFH unit was designed with tankage for a 10 hour mission; the 10 SCFH unit was designed with tankage for a 10 hour mission and, as an alternate, with tankage for a 2 hour mission.

The mechanical design and operation of a 4 SCFH hydrogen generator is illustrated schematically in Figure 20. The operation of the unit is as follows:

Fuel and steam flow from their respective tanks, are metered, pre-mixed and pass through the vaporizer-preheater coil before entering the reformer. Flue gases passing over the finned underside of the tank transfer sufficient heat to generate the steam. The liquid fuel is pressurized by means of a flexible diaphragm between the fuel and steam chambers.

Flue gases passing over the vaporizer-preheater coil supply the heat to complete the vaporization and preheat the fuel steam mixture to reactor entry temperature. The fuel-steam mixture enters the reactor where in the presence of steam reforming catalyst and utilizing heat transferred from the combustion zone, it is reformed to carbon dioxide, carbon monoxide, hydrogen and methane; excess water is present. The product hydrogen is removed through the diffusion element and passes to the hydrogen delivery line through a final cooling coil.

The residual fuel gas enters the operational burner where it is burned. Air is aspirated into the combustion zone by the pressure of the residual fuel gas passing through the orifice which maintains the system pressure. The combustion products flow through the reactor, the preheating coil, steam generator and out the exhaust.

The 10 SCFH hydrogen generator is shown in Figure 21. A 10 SCFH breadboard model was constructed in favor of the 4 SCFH unit for delivery to the U.S. Army Electronics Laboratory. The steam boiler pressurization method used for the 4 SCFH design was found to be unsuitable for this size because of less favorable heat transfer surface to container volume. Also, the mass of the boiler and its water content increases to the extent that it would require a prohibitively long time to heat them to start-up temperature. Instead, a compressed air pressurization system is used. In the absence of a feed-water boiler a readily replaceable vaporizer-preheater, located in the upper portion of the combustion tube, has been provided to vaporize the fuel and water and act as an accumulation point for minerals dissolved in the water. Gum and other fuel deposits will also collect at this point. Except for these changes, the operating characteristics of the 10 SCFH

hydrogen generator are similar to the 4 SCFH unit.

The start up of either unit is accomplished by burning liquid hydrocarbon fuel in a blowtorch type start-up burner, providing heat to bring the reactor body to start-up temperature. Water and fuel are then forced into the reactor, initiating the reforming reaction and providing a residual fuel gas to the operational burner which is ignited by the flame from the start-up burner.

When substantial hydrogen product gas is being produced, which is estimated to be within 20 minutes after initial start-up, the liquid fuel supply to the start-up burner is shut off, and the unit is on stream and self-sustaining.

The necessary conditions to deliver the desired quantity of hydrogen are that the diffusion element be properly sized and that an adequate hydrogen partial pressure difference exists across it. With a properly sized diffusion element, the hydrogen partial pressure difference is obtained by maintaining a sufficiently high total reactor pressure by means of a calibrated fixed area orifice in the reactor outlet line. Reactor pressures are on the order of 150 psig. With the reactor pressure fixed in this manner, a sufficient flow of fuel and the proportionate flow of water to maintain the reactor pressure are admitted into the unit by controlling the pressure drop across calibrated fixed area orifices.

The quantity of hydrogen delivered by this generator can be manually throttled by reducing the pressure drops across the feed orifices. With less material flowing into the reactor and through the effluent orifice, a lower reactor pressure and hence a lower hydrogen delivery rate results. Theoretically, this could be continued until no net hydrogen production results. Practically, the reduction of the hydrogen production is limited. The reason for this is that the gases leaving the reformer back-pressure orifice provide the motive force for aspirating the combustion air. At low flows, not enough combustion air would

be aspirated. Data at the present time leads us to believe that the lower limit may be approximately 75% of full flow.

During any nominally constant operation of a fuel cell power supply and its hydrogen generator, small transient fluctuations of operating conditions will occur. Without control provisions, a momentary drop in hydrogen delivery or rise in hydrogen demand would result in a decrease in delivery line pressure. Conversely, a momentary rise in hydrogen delivery or drop in hydrogen demand would cause a rise in line pressure. In larger fully instrumentated units corrective action is provided. In small units, these fluctuations are allowed for by delivering a quantity of hydrogen slightly in excess of that required as an average by the fuel cell at this load and venting this excess whenever the hydrogen delivery line pressure rises.

Results of the design studies have provided the size and weight limitations shown in Table 18.

4. Component Studies

The design studies pointed out the need for miniature, lightweight components. Laboratory evaluations of several of these components were undertaken to study their suitability and reliability for this application.

It was found that many miniature, lightweight components were commercially available with operating characteristics which met our requirements. These included pressure regulators, pressure relief valves, filters, pressure gauges, block valves, flowmeters, and insulating materials. Items which were not available and which had to be developed were as follows:

a. Metering Orifices

Several commercial miniature metering valves were purchased and tested in the laboratory. The test equipment is shown schematically in Figure 22. It was found that none of the valves gave satisfactory operation at the required extremely low flow rates. This has been attributed, in the main, to stem backlash. As an alternate to commercial

metering valves, miniature sharp edged orifices have been designed, fabricated and tested. Results indicate that these orifices perform satisfactorily.

b. Fuel, Water, and Air tanks

Various methods of fabrication, materials of construction, and tank geometries were investigated so as to provide minimum weight and over-all volume and ease of draining. Materials considered included spun aluminum, spun stainless steel, and fiberglass-reinforced plastics. Fiberglass-reinforced-polyester tanks obtained from one vendor to evaluation were found to be unsuitable as they failed a hydrostatic pressure test at or below design pressure. These tanks, however, were produced by Hand-Lay-Up methods and their failure does not preclude the suitability of precision filament-wound tanks.

c. Air Compressor

A survey was made to determine the availability of a small light weight hand-operated air compressor, capable of compressing air from atmospheric pressure to 300 psig. As no commercial model could be obtained, a program was undertaken to design, develop, and test a compressor. The feasibility of using a single stage piston-operated compressor was first established using specially designed linkages with commercial pneumatic cylinders. It was then established that commercial cylinders (such as aluminum or plastic) could not practically be modified for use either because of their weight or because too much dead volume existed at the end of the piston stroke so that the compression ratio was too low.

A prototype aluminum hand-operated compressor was designed which employed a one piece cylinder and head of high strength aluminum alloy 6061. An aluminum piston was designed which incorporated an O-ring dynamic seal. The pump bore was 1.5 inches, with a stroke of 2 inches. The miniature check valves were modified, by reducing their length, so as to reduce the pump dead volume. Suitable linkages and framework were designed and the pump was fabricated. The weight of the assembled unit was 2.2 pounds.

Testing indicated that the pump was capable of compressing atmospheric air to pressures in excess of 300 psig. A 55 cubic inch air tank would supply pressure for about 1 hour of generator operation. The time to pressurize the tank from atmospheric pressure to 300 psig was 12 minutes and to re-pressurize from 160 to 300 psig was 7 minutes, both at a rate of about 50 strokes per minute. Improvements, in future models, to reduce the time to pressurize the air tank could include a longer piston stroke or consideration of a double-acting piston that would compress air at each end of the cylinder.

5. Experimental Hydrogen Generators

An experimental liquid hydrocarbon fueled hydrogen generator, designated PT-8, was designed, fabricated, and tested. For initial operating tests, Whitey Laboratory Feed Pumps were used for close control of the feed streams. Initial operation of the unit indicated problem areas existed with the start-up burner (insufficient heat release); and with the operational burner (instability and excessive back pressure).

Modifications were made and the unit was operated at production rates of up to 10 SCFH at 10 psig delivery pressure (for a gross heating value efficiency of 52%). Operation at 10 SCFH hydrogen delivery rate and low hydrogen delivery pressure was unstable, and start-up time was in excess of 60 minutes due to insufficient heat release from the start-up burner. The test was discontinued after 240 hours of operation.

At this point sufficient information had been obtained to design and fabricate a more compact hydrogen generator, PT-9 for use in the breadboard assembly. This generator was test operated prior to delivery of the breadboard assembly. The generator was successfully operated on a test stand using laboratory feed pumps. The unit was then integrated with the separately developed pressurized air feed system and the complete assembly was mounted on a panel as a breadboard for delivery to Fort Monmouth. The system was first operated by starting with pumps and then switching to the pressurized feed system, and; finally, was started up and

operated with the air pressurized feed system. Start-up was accomplished within 30 minutes.

Typical performance data for the PT-9 hydrogen generator is shown in table 19. Hydrogen production rates of 11 to 14.5 SCFH were obtained with gross heating value efficiencies of 52 to 71%. During test periods A and B the generator was operated on a test stand using laboratory feed pumps; the hydrogen production rate was 11 to 11.7 SCFH, and the gross heating value efficiency was 52 to 54%. During test period C the generator was operated by starting with laboratory feed pumps and then switching to the pressurized feed system; the hydrogen production rate 10.8 to 13 SCFH, and the gross heating value efficiency was 53 to 64%. During test period D the generator was started up and operated with the air pressurized feed system; the hydrogen production rate was 14.5 SCFH, and the gross heating value efficiency was 71%.

On October 20, 1964 the breadboard liquid hydrocarbon fueled hydrogen generator, PT-9, was delivered to Fort Monmouth and was set up for a demonstration. After clearing a partial blockage that occurred, the unit was successfully operated and produced in excess of rated capacity. A flow diagram of this unit is shown in Figure 24 and a photograph is shown in Figure 25.

When operating the integrated breadboard assembly, it was found that instabilities sometime occurred in the system; i.e., large fluctuations in reactor pressure with occasional flameout or burning outside of the reactor at the top of the stack.

The fuel cost per KWH based upon liquid hydrocarbon fuel in tankwagon quantity at 20.5 ¢/gal. has been calculated to be 17 mils/KWH based on a hydrogen consumption of 10 SCFH by a 500 watt fuel cell.

V CONCLUSIONS

Investigations have shown that it is possible to generate ultra-pure hydrogen for fuel cell use on a portable basis. Fuels considered include ammonia, methanol, and liquid hydrocarbons.

Several versions of an ammonia fueled hydrogen generator have been constructed and tested. Mechanical failures that occurred provided information for design improvements such as use of a bellows assembly to compensate for thermal stresses.

A final prototype portable ammonia feed hydrogen generator substantially meeting the contractual requirements specified has been delivered to the U.S. Army Electronics Laboratory. This unit is capable of delivering various outputs of hydrogen depending on ammonia feed pressure and is completely self-sustaining. Thermal efficiencies of over 70% are possible with the generator.

Tests were performed with ammonia fueled generators to obtain a life study and to improve and modify procedures and ancillary equipment. Test generator PT-7 was operated for 490 hours before a mechanical failure occurred at a welded joint. Test generator PT-6 completed 87 cycles for 1035 hours of operation at full rated capacity before failure. The failure was caused by nitride formation in conjunction with mechanical and thermal stresses. Latest design incorporates a higher nickel alloy reactor tube to resist nitride formation and bellows to compensate for thermal stresses caused by differential thermal expansion.

Experimental work with methanol has shown that it can be reformed with an equimolar ratio of water with hydrogen production such that a portable unit utilizing its own off-gas for fuel could be developed to yield the design hydrogen flow.

Experimental work with liquid hydrocarbon feeds showed that they could be steam reformed to produce hydrogen at thermal efficiencies such that a portable unit using its own off-gas for fuel could be developed.

Results of catalyst life tests indicated liquid hydrocarbons such as BTX UDEX Raffinates or JP-4 jet fuel could be

successfully steam reformed at molar water to carbon atom ratios of 3 with reasonable catalyst life provided the sulphur content was below 40 PPM.

Detailed design studies were performed to design units consuming selected liquid hydrocarbons at the 4 SCFH level (200 watt) and at the 10 SCFH level (500 watt). Results of the design studies have provided the following estimate of size and weight limitations. For a 4 SCFH unit with tankage for 10 hours a size of 2 cubic feet and a dry weight of 20 pounds; for a 10 SCFH unit, a volume of 2 cubic feet and a dry weight of 25 pounds based upon a 2 hour mission or a dry weight of 30 pounds based upon a 10 hour mission.

Results of component studies indicated that lightweight components meeting operating requirements could be obtained or fabricated.

Two experimental liquid hydrocarbon fueled hydrocarbon generators, (PT-8 and PT-9) were designed, fabricated, and tested. Unit PT-8 was operated for 240 hours with hydrogen production rates of up to 10 SCFH at 10 psi delivery pressure for a gross heating value efficiency of 52%. Unit PT-9 was operated at hydrogen production rates of 11 to 14.5 SCFH with gross heating value efficiencies of 52 to 71%. Start up was accomplished within 30 minutes.

The PT-9 hydrogen generator was integrated with a hand operated air-pressure feed system assembled on a panel as a breadboard model and delivered to the U.S. Army Electronics Laboratory, Fort Monmouth, where a demonstration run was performed.

Certain instabilities remain in this generator which require further work in the area of component and systems optimization in order to provide a field type hydrogen generator.

Based upon a liquid hydrocarbon fuel cost of 20.5¢/gal. in tankwagon quantity, a primary fuel cost of 17 mils/KWH has been established.

VI. IDENTIFICATION OF KEY TECHNICAL PERSONNEL
AND DISTRIBUTION OF HOURS

The following technical personnel have been working on the
hydrogen generator for the period 31 January - 31 October, 1964:

L. Goodman	1439
R. Gaunt	1033
W. Muldowney	221
H. Hess	615
R. Maloney	410
C. Freeman	32
Miscellaneous Engineers	16
Drafting	300
Technicians	<u>2329</u>
	6395 hours

APPENDIX A

Tables

TABLE 1

SUMMARY OF DATA FOR AMMONIA FUEL

Portable Hydrogen Generator

Run No.	Orif. Diam. (mils) ¹	Prod. H ₂ (SCFH) ²	Tot. H ₂ in Prod. (SCFH)		Ammonia Feed (SCFH)		N ₂ Stack Gas (SCFH sat. 32°)	% H ₂ in Fuel Gas	Matl. Eff. (lb. H ₂ /lb. NH ₃)	Therm. Eff. (GHV H ₂ /GHV NH ₃)
			Meas. ³	Calc. ⁴	Meas. ⁵	Calc. ³				
1	12.6	3.81	7.18	-	-	4.79	8.02	58.4	0.092	61.8
2	12.6	3.70	6.82	-	-	4.54	8.10	57.8	0.095	63.9
4	13.0	4.58	8.98	-	-	5.99	10.69	59.5	0.089	59.8
6	13.0	4.30	7.99	7.65	5.10	5.30	9.10	57.7	0.097	65.5
7	13.0	4.10	7.61	7.65	5.13	5.08	9.10	58.7	0.093	62.3
8	13.0	4.40	8.20	7.76	5.08	5.46	8.68	57.6	0.099	66.5
9	13.0	4.65	8.16	7.58	5.05	5.44	9.15	55.1	0.102	69.0
10	13.0	4.60	8.06	7.35	4.90	5.38	8.83	54.6	0.104	69.7
11	13.0	4.50	8.11	7.58	5.05	5.40	8.63	56.1	0.100	67.1

1. Throat diameter of aspirator assembly was 0.073" for all runs.
2. Unit was run with 1 psig back pressure to simulate feed conditions necessary to operate actual fuel cell (except Run 1 which was run at atmospheric pressure).
3. From H₂O condensate, product H₂, and stack gas analysis.
4. From measured NH₃ feed.
5. Rotameter and/or weight fed.
6. GHV = Gross Heating Value.

TABLE 2

SUMMARY OF DATA FOR AMMONIA FUEL

Portable Hydrogen Generator

Reactor Designation	Cycle No.	Product H ₂ (SCFH)	Total H ₂ in Prod. (SCFH)	Ammonia Feed (SCFH)	% H ₂ in Fuel Gas	Mat'l Eff. (lb H ₂ /lb NH ₃)	Therm. Eff. * (GHV H ₂ /GHV NH ₃)
PT-3	13	4.2	7.4	4.9	57.1	0.099	66.6
	16	4.0	7.2	4.8	57.1	0.097	65.2
	20	4.3	7.2	4.8	54.7	0.104	69.8
PT-5 (Parallel Flow)	2	1.9	6.2	4.1	68.3	0.054	36.3
PT-5 (Counter-current Flow)	2	3.8	7.8	5.2	60.6	0.085	57.1
	3	4.0	7.95	5.3	59.8	0.088	60.2
	4	3.5	6.6	4.4	58.5	0.092	61.8

* Thermal Efficiency = Gross Heating Value Product H₂/Gross Heating Value NH₃

TABLE 3

SUMMARY OF DATA FOR AMMONIA FUEL

Portable Hydrogen Generator--PT-3A

Cycle No.	Product H ₂ (SCFH)	Total H ₂ in Prod. (SCFH)	Ammonia Feed (SCFH)	% H ₂ in Fuel Gas	Mat'l. Eff. (lb H ₂ /lb NH ₃)	Therm. Eff. * (GHV H ₂ /GHV NH ₃)
1	4.5	7.6	5.05	55.4	0.103	69.2
3	4.7	8.1	5.4	55.7	0.101	67.8
6	4.8	8.55	5.7	56.8	0.098	65.8
8	4.5	7.8	5.2	55.9	0.100	67.2
10	4.4	7.35	4.9	54.6	0.104	69.8

* Thermal Efficiency = Gross Heating Value H₂/Gross Heating Value NH₃

TABLE 4

SUMMARY OF DATA FOR AMMONIA FUEL

Portable Hydrogen Generator

Reactor Desig.	Cycle No.	Product H ₂ (SCFH)	Total H ₂ in Prod. (SCFH)	NH ₃ Feed (SCFH)	% H ₂ in Fuel Gas	Mat'l Eff. (lb H ₂ /lb NH ₃)	Therm. Eff. * (GHV H ₂ /GHV NH ₃)
PT-3A	11	4.0	7.20	4.8	57.1	0.097	64.9
PT-3B	1	1.1	7.35	4.9	71.8	0.026	17.6
	2	3.1	7.80	5.2	64.5	0.069	46.5
	5	2.3	6.75	4.5	66.4	0.059	39.8
PT-3C	1	3.8	7.70	5.15	60.0	0.085	57.2
	3	4.0	7.70	5.15	58.8	0.090	60.5
	6	4.2	7.70	5.15	57.4	0.095	63.8
PT-3D	1	4.0	7.60	5.05	59.1	0.092	61.3
	2	3.9	7.50	5.0	59.0	0.090	60.8

* GHV = Gross Heating Value

TABLE 5

SUMMARY OF DATA FOR AMMONIA FUEL

Portable Hydrogen Generator - PT-6

Cycle No.	Product H ₂ (SCFH)	Total H ₂ Produced (SCFH)	NH ₃ Feed (SCFH)	% H ₂ in Fuel Gas	Mat'l. Eff. (lb H ₂ /lb NH ₃)	Therm. Eff.* (GHV H ₂ /GHV NH ₃)
1 ¹	4.35	8.78	5.85	60.0	0.086	57.8
4	4.40	8.93	5.95	60.3	0.086	57.7
7	4.10	8.33	5.55	60.5	0.086	57.6
12	4.10	8.29	5.53	60.3	0.086	57.7
16	4.00	8.29	5.53	60.9	0.084	56.5
20	4.50	9.31	6.20	61.0	0.084	56.3

1401

* Thermal Efficiency = Gross Heating Value of Product H₂/Gross Heating Value of Ammonia Feed.

1 Reactor operating pressure 35 psig for this run; remaining runs were performed at 30 psig.

TABLE 6
SUMMARY OF COMPONENT WEIGHTS
SAMPLE HYDROGEN GENERATOR
AMMONIA FUELED

<u>Component</u>	<u>Weight</u>
Reactor Assembly	4 lb. 3 oz.
NH ₃ Container	2 lb. 14 oz.
NH ₃ Charge	2 lb. 12 oz.
Cannister	1 lb. 8 oz.
Insulation	3 lb. 9 oz.
Battery + Switch	3 oz.
Gauges + Regulator + Fittings	<u>1 lb. 14 oz.</u>
	13 lbs. 63 oz. =
	16 lbs. 15 oz.

TABLE 7

SUMMARY OF DATA FOR AMMONIA FUEL

Portable Hydrogen Generator - PT-7A

Cycle No.	Product H ₂ (SCFH)	Ammonia Feed (lb/hr.)	Mat'l Eff. (lb H ₂ /lb NH ₃)	Therm. Eff.* (GHV H ₂ /GHV NH ₃)	Start-up Time to Design H ₂ Flow (Mins.)
2	4.5	.243 ¹	0.0978 ¹	65.7	35
3	4.3	.208	0.109	73.4	30
5	4.6	----	-----	-----	30

* Thermal Efficiency = Gross Heating Value of Product H₂ / Gross Heating Value of Ammonia Feed.

¹ Unit was run at pressures between 35 psig and 25 psig for varying intervals. Figures indicate magnitude of hydrogen production.

TABLE 8

SUMMARY OF DATA FOR AMMONIA FUEL

Sample Hydrogen Generator Device-PT-7

Cycle No.	Product H ₂ (SCFH)	Ammonia Feed (FT ³ /HR) ¹	(lb/HR) ²	Mat'l Eff. (Lb H ₂ /Lb NH ₃)	Therm. Eff. ³ (GHV H ₂ /GHV NH ₃)
1	4.25	4.6		0.107	71.9
4	4.15		0.250	0.088	59.9
5	4.25		0.226	0.101	67.8
9	4.20	4.6		0.106	71.2
13	4.10		0.256	0.085	57.2
19	4.15	4.7		0.102	68.6
23	4.25		0.276	0.081	54.4

1. Feed from the ammonia cylinder and measured by rotameter.
2. Average feed rate when using the portable bomb as determined by weight.
3. Thermal Efficiency = Gross Heating Value of Product H₂/Gross Heating Value of Ammonia Feed.

TABLE 9

SUMMARY OF DATA AMMONIA FUEL

Sample Hydrogen Generator - PT-7

Cycle No.	Product H ₂ (SCFH)	Ammonia Feed (1) (Ft ³ /Hr.)	Mat'l Eff. (lb H ₂ /lb NH ₃)	Therm. eff. (2) (GHV H ₂ /GHV NH ₃)
25	4.1	4.75	0.100	67.2
29	4.2	4.8	0.102	68.7
32	4.2	4.75	0.103	69.2
35	4.1	4.8	0.099	66.5
39	4.2	4.75	0.103	69.2

(1) Feed rate as measured by rotameter.

(2) Thermal Efficiency = Gross Heating Value of Product H₂/Gross Heating Value of Ammonia Feed.

TABLE 10

SUMMARY OF DATA FOR AMMONIA FUEL

Hydrogen Generator - PT-6

Cycle No.	Product H ₂ (SCFH)	Ammonia Feed ¹ Ft ³ /Hr.	Mat'l Eff. (lb H ₂ /lb NH ₃)	Therm. Eff. ² (GHV H ₂ /GHV NH ₃)
21	4.2	5.1	0.096	64.6
29	4.2	5.2	0.094	63.3
36	4.2	5.1	0.096	64.6
43	4.2	5.3	0.092	61.3
51	4.2	5.4	0.090	60.5
59	4.15	5.4	0.089	59.8

1. Feed rate as measured by rotameter.

2. Thermal Efficiency = Gross Heating Value of Product H₂/Gross Heating Value of Ammonia Feed.

TABLE 11

SUMMARY OF DATA FOR AMMONIA FUEL

Hydrogen Generator - PT-6

Cycle No.	Product H ₂ (SCFH)	Ammonia Feed ¹ (SCFH)	Mat'l. Eff. (lb.H ₂ /lb.NH ₃)	Therm. Eff. ² (GHV H ₂ /GHV NH ₃)
60	4.15	5.3	0.091	61.1
65	4.10	5.3	0.090	60.5
70	4.18	5.1	0.095	63.8
75	4.20	5.0	0.097	65.2
80	4.30	5.2	0.096	64.5
85	4.10	5.15	0.092	61.8
87	4.40	5.4	0.095	63.8

1 Feed Rate as Measured by Rotameter.

2 Thermal Efficiency = Gross Heating Value of Product Hydrogen/
Gross Heating Value of Ammonia Feed.

TABLE 12
SUMMARY OF DATA FOR METHANOL FUEL

<u>Operating Conditions</u>	<u>First Period</u>	<u>Second Period</u>
TIME-HOURS	13	55-1/2
FEED RATES		
Water, CC/Hr.	400	390
Methanol, CC/Hr.	900	890
Vol. Ratio - H ₂ O/CH ₃ OH	.44/1	.44/1
Molar Ratio - H ₂ O/C	1/1	1/1
REACTOR		
Temperature, °F	1000	1000
Space Velocity (CH ₃ OH) (Gms. of feed/Hr./Gm.Cat.)	1.15	1.15
Pressure, psig	150	150
RESULTS		
Product Gas, CF/Hr.	39	44
Hydrogen, CF/Hr.	28	31
Catalyst Loading Gms. of CH ₃ OH/Gm.	15	63.5

TABLE 13

SUMMARY OF DATA

Steam Reforming of JP-4 Fuel

Run Hour No.	9	16.5	45.5
cc Oil/hr. (S.G. HC = 0.767)	60	50	63
Pure Hydrogen Production (SCFH)	4.62	4.84	5.80
Mat'l Efficiency (SCFH H ₂ /100 cc HC)	7.70	9.68	9.21
Off-Gas Analysis (Dry Basis, Mol%)			
Methane	8.1	7.5	---
CO ₂	38.9	41.3	---
CO	26.2	27.9	---
H ₂	27.2	22.3	---
Carbon Balance (%)	89.0	107.1	---
Hydrogen Balance (%)	101.3	103.4	---
Thermal Efficiency (%)*	73.8	93.0	88.3

* Gross Heating Value of Product Hydrogen/Gross Heating Value of Feed.

TABLE 14
EMULSIFIER SCREENING TESTS¹

JP-4 - Water

<u>Emulsifier</u>	<u>Commercial Source</u>	<u>Results</u>
Tergitol 7	Union Carbide	Complete Separation
" 14	" "	" "
" NP-27	" "	" "
" NPX	" "	" "
Tergitol 14+ Tergitol NP-27	" "	" "
" 14+ " NPX	" "	" "
" NP-27+ " NPX	" "	" "
Triton X- 45	Rohm and Haas	" "
" X- 100	" "	" "
" GR-7	" "	" "
Triton 45+)	" "	Stable
" 100+)Mixture	" "	for
" GR-7)	" "	Few Hours
Triton N-57	" "	Complete Separation
9D-208	" "	Stable for 100 Hours
Petromix 10	Sonneborn Corp.	Stable for 100 Hours
Petronate HL	" "	Complete Separation

- 1 These emulsifiers were selected after contacting various companies and reviewing their recommendations in regard to the conditions and requirements of a successful emulsion.

TABLE 15

INSPECTION BTX - UDEX RAFFINATE

A. Distillation ASTM D-86

Initial Boiling Point	166°F
10% by volume	180
20	184
30	189
40	193
50	199
60	207
70	215
80	228
90	249
End Point	277
Recovery	97.5%

B. Specific gravity 60°F/60°F = 0.6991; hence 70.9° API

C. Sulphur 1ppm

TABLE 16

INSPECTION JP-4

A. DISTILLATION ASTM D-86

Initial Boiling Point	140°F
10%	196
20	216
30	233
40	248
50	264
60	282
70	308
80	332
90	372
End Point	433
Recovery	98.0%

B. Specific gravity 60°F/60°F = 0.7686, hence 52.6° API

C. Sulphur 42 ppm

TABLE 17

EQUILIBRIUM CALCULATIONS

Hour of Run	500	620	641	668	933	979
CO Reform Equil. Temp., °F	1502	1497	1466	1459	1446	1448
CO ₂ Reform Equil. Temp., °F	1504	1497	1453	1462	1446	1435
Shift Equil. Temp., °F	1552	1527	1570	1516	1484	1570

Catalyst operating temperature 1500°F in all cases.

TABLE 18

DESIGN STUDY RESULTS

	Overall Volume Cubic Feet	Weight, Pounds		Fuel & H ₂ O Requirement Pounds/Hour
		2 Hour Mission	10 Hour Mission	
4 SCFH Unit (200 Watt) Self Pressurized	2.0	X	20	0.6
10 SCFH Unit (500 Watt) Air Pressurized	2.0	25	30	1.5

TABLE 19

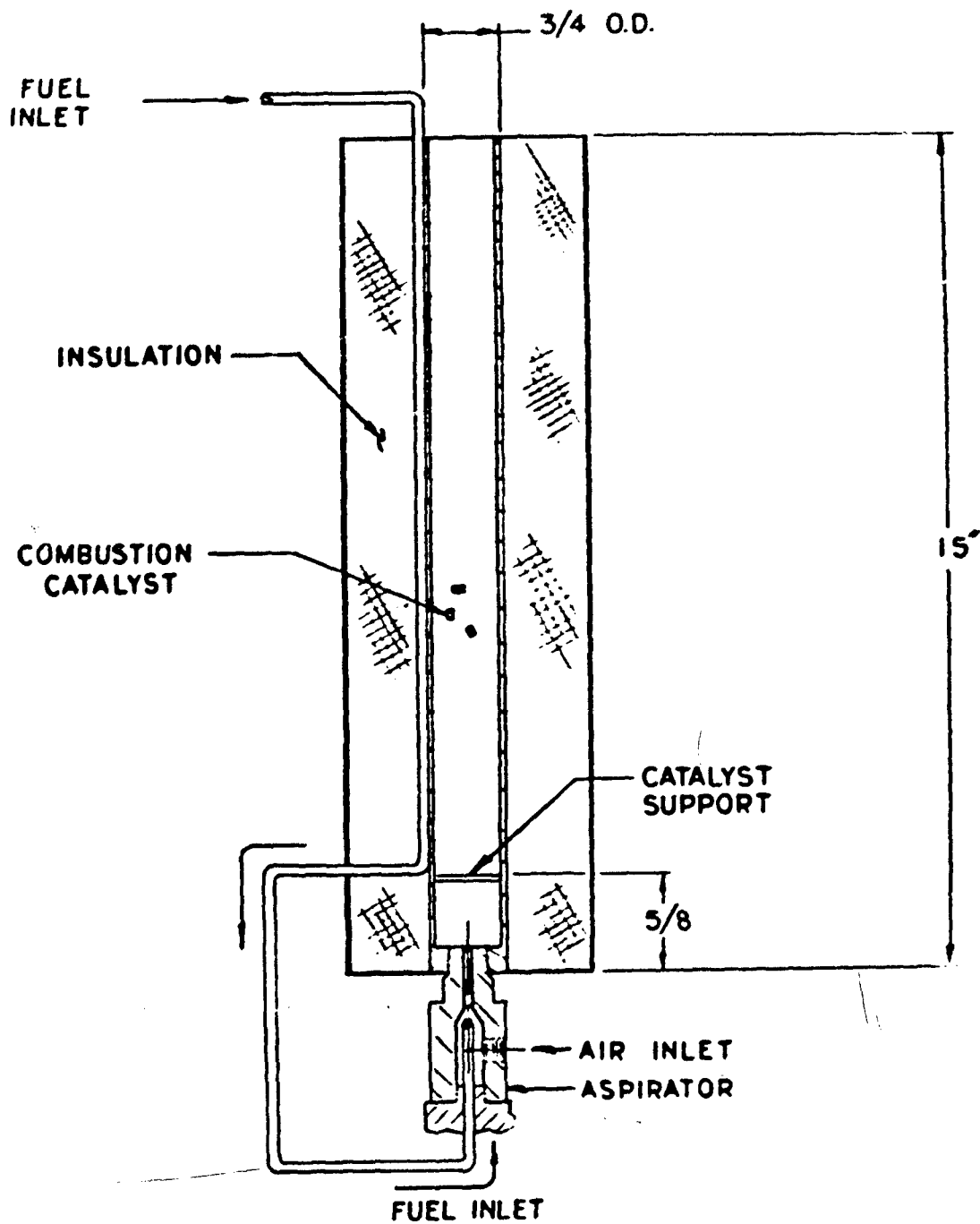
SUMMARY OF TEST DATA

Hydrogen Generator PT-9

Test Period	Test Duration Hours	H ₂ Production Rate (SCFH)	Hydrogen Delivery Pressure, psig	Water/carbon Molar Ratio	Gross Heating Value Eff. %
A	14	11.0	5.0	2.5	52
B	26	11.7	1.5	2.5	54
C	2.5	10.8 to 13.0	6.0	2.5	53-64
D	1	14.5	1.0	2.8	71

APPENDIX B

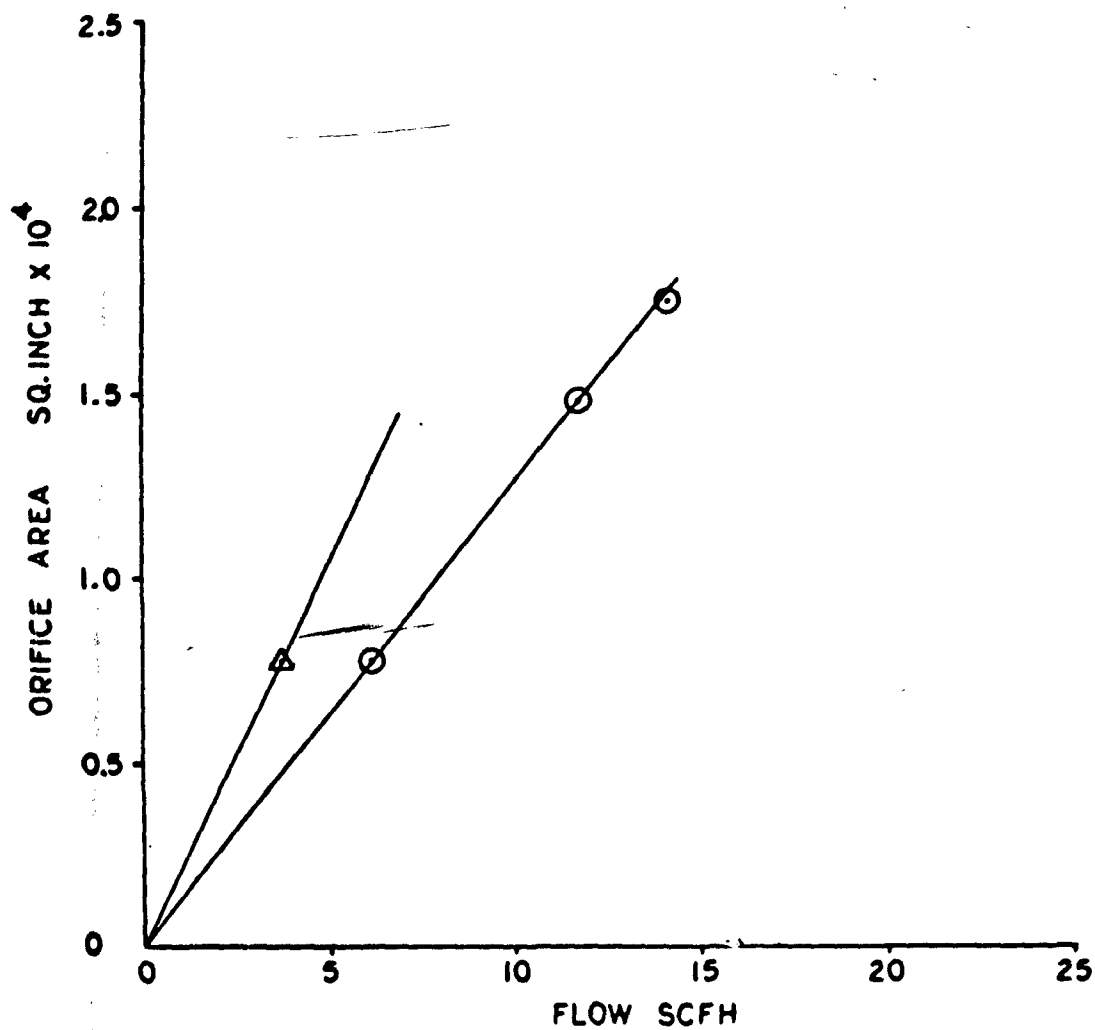
Figures



COMBUSTION CHAMBER

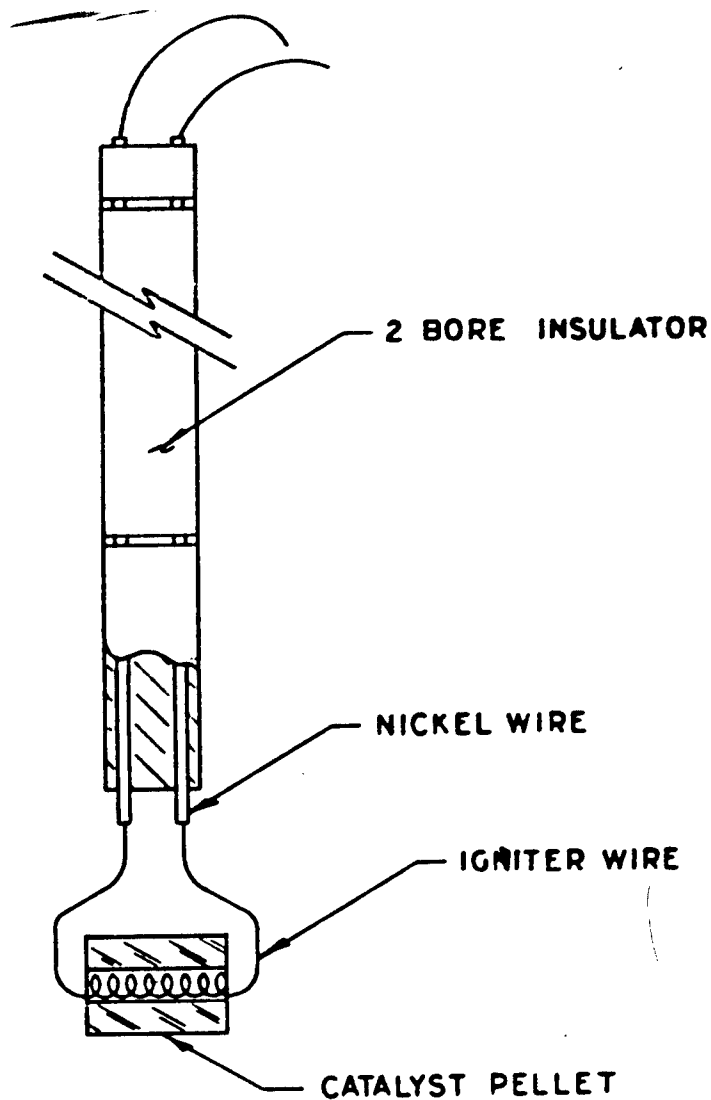
FIG. 1

○ - ROOM TEMP
△ - OPERATING TEMP



FLOW VS ORIFICE AREA

FIG. 2



IGNITER

FIG. 3

FIG. 4
IGNITER SYSTEM

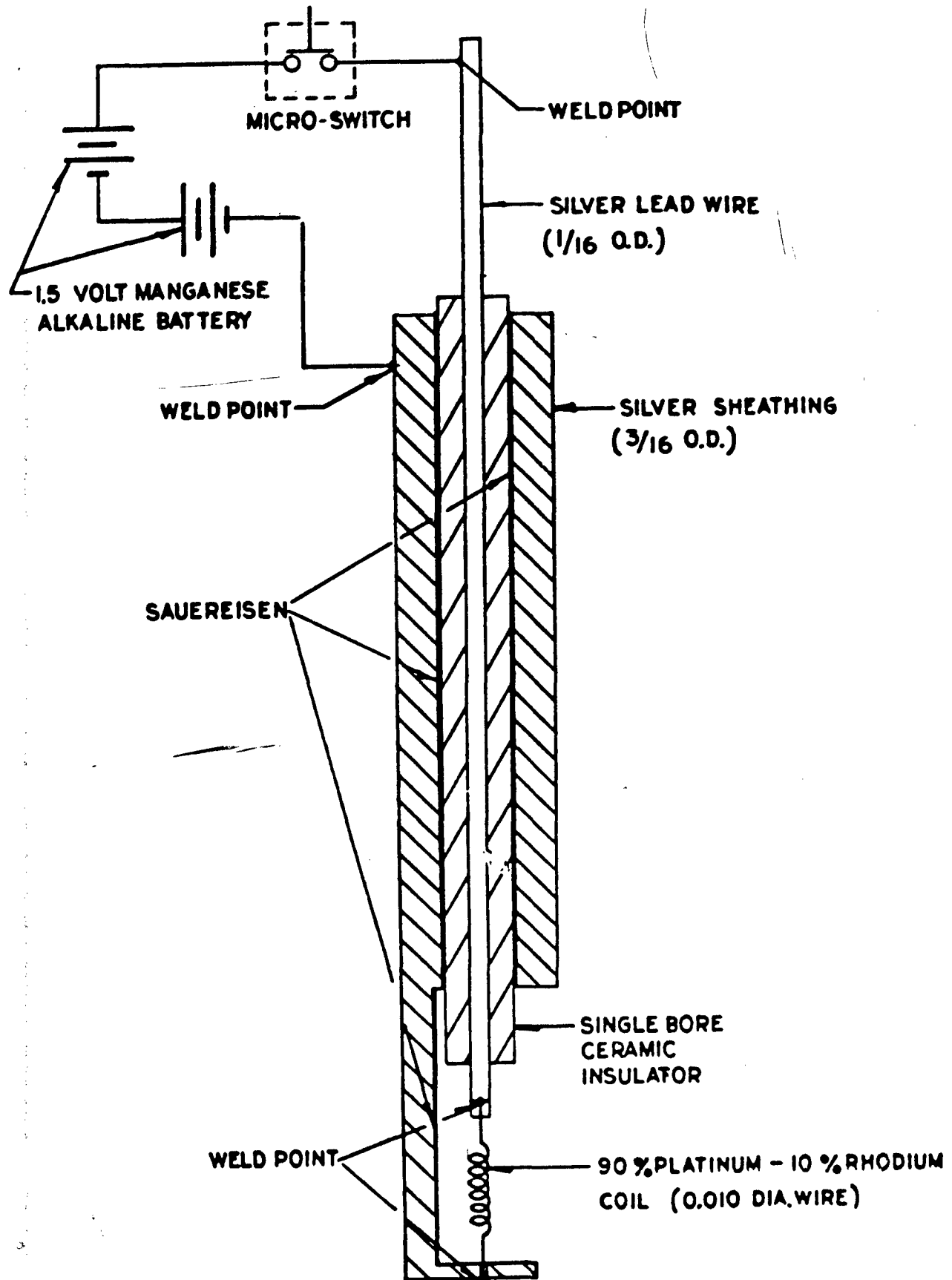


FIG. 5
SCHEMATIC FLOW DIAGRAM-TEST SETUP

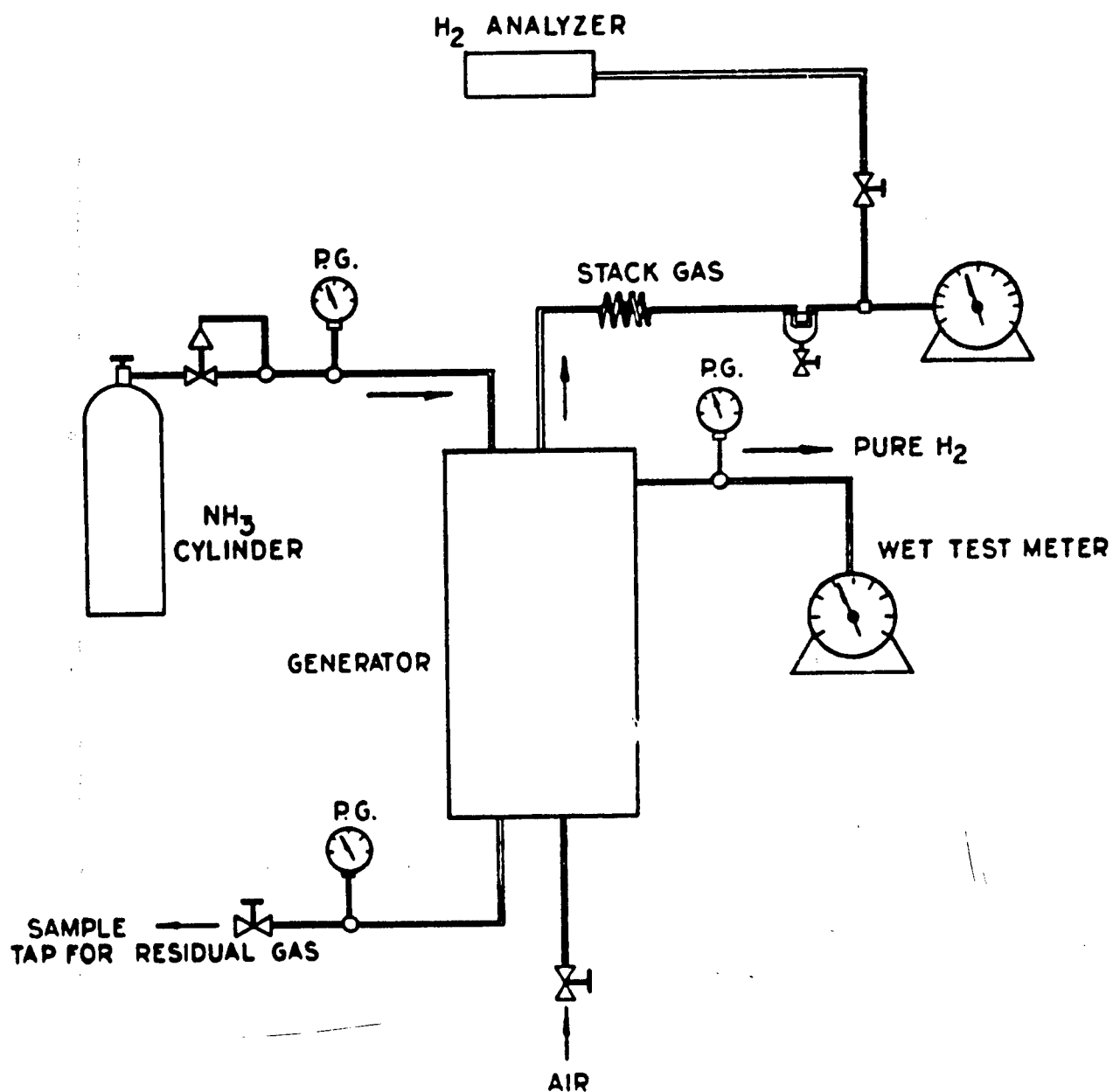


FIG. 6
SCHEMATIC FLOW DIAGRAM
TEST SET UP TYPICAL
EXPERIMENTAL PORTABLE H₂ GENERATOR
AMMONIA FUELED

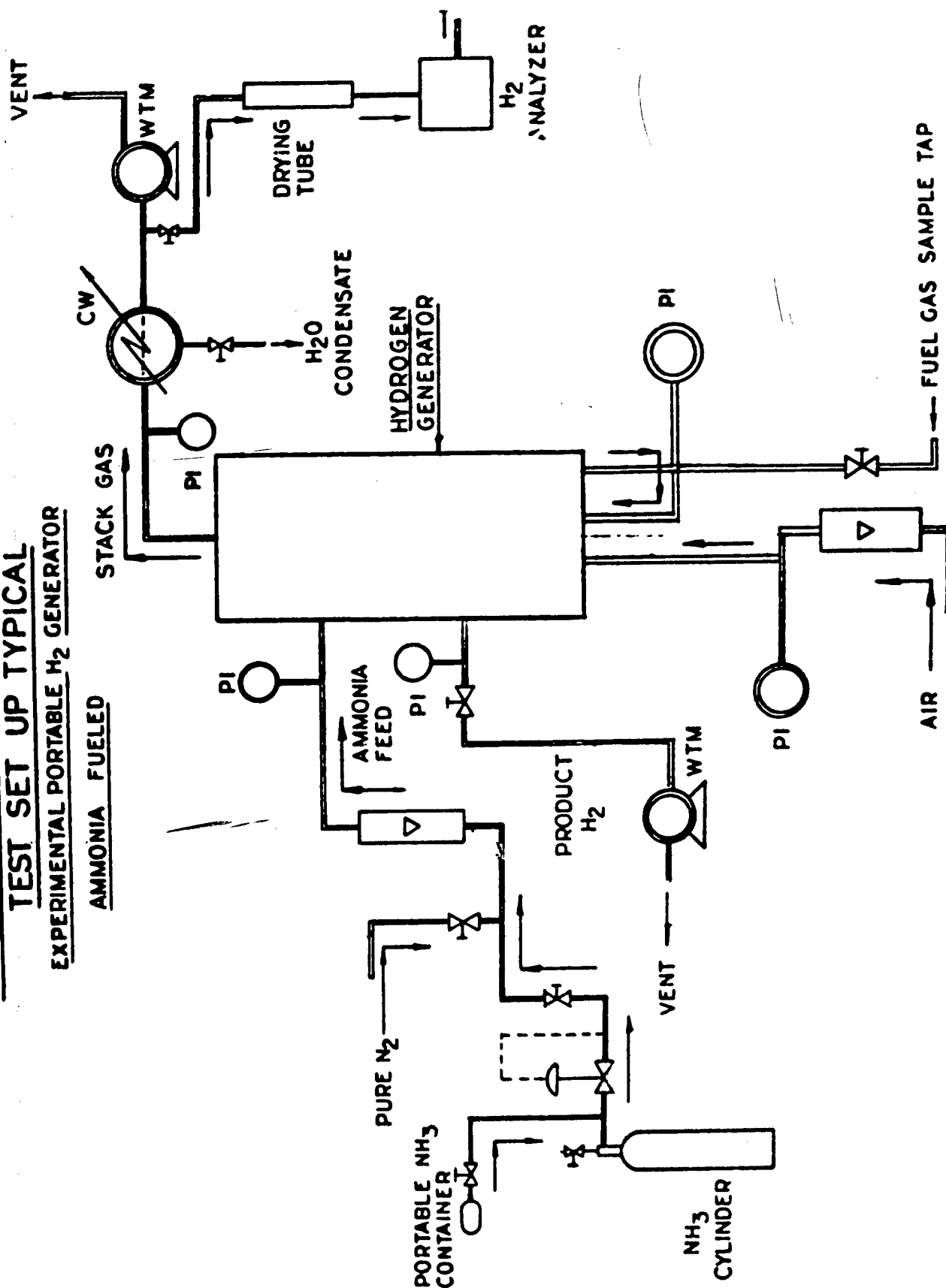


FIG. 7
TEMPERATURE PROFILE
PORTABLE H₂ GENERATOR
PT-3

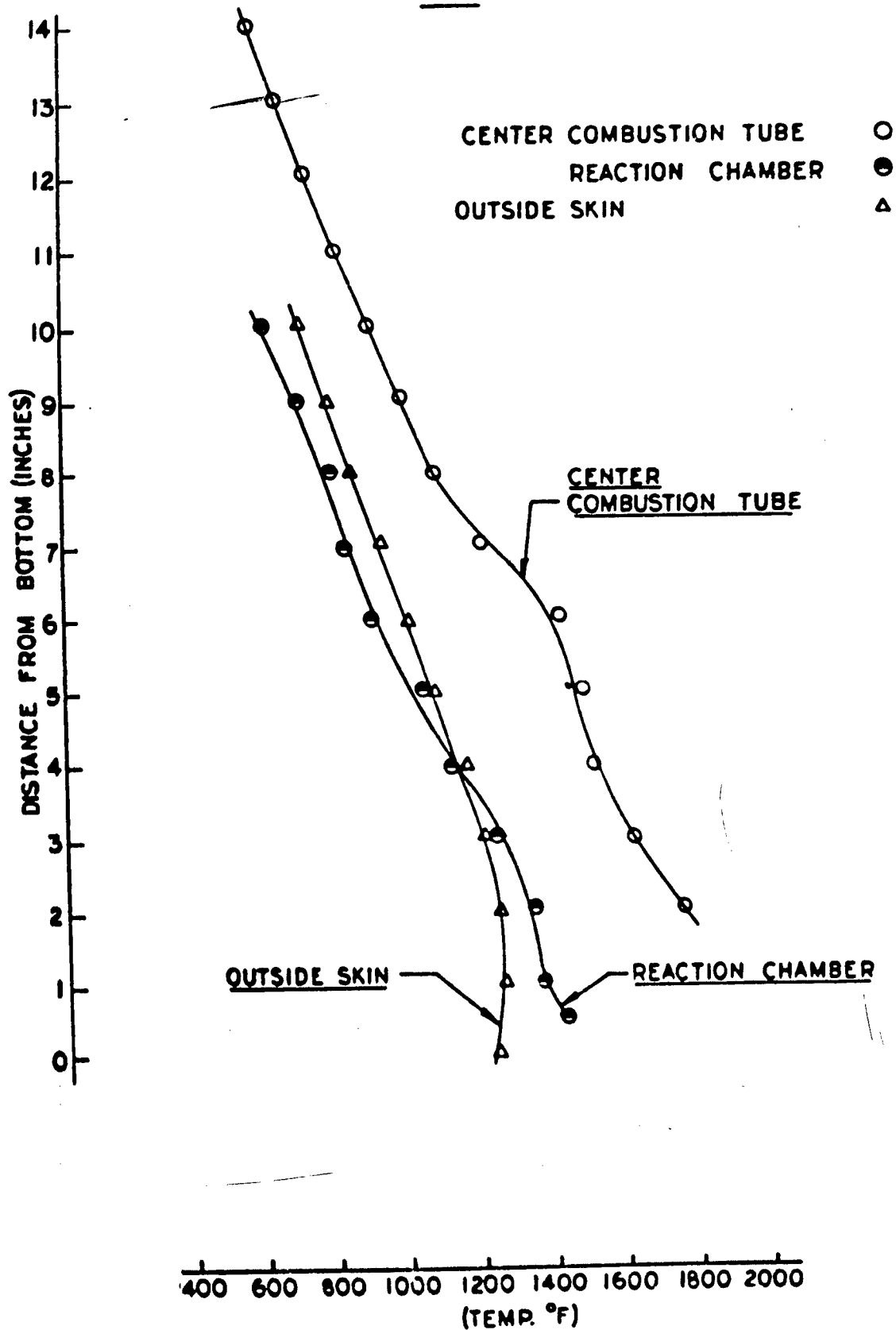


FIG. 8
TEMPERATURE PROFILE
PORTABLE H₂ GENERATOR
PT-5

PARALLEL FLOW

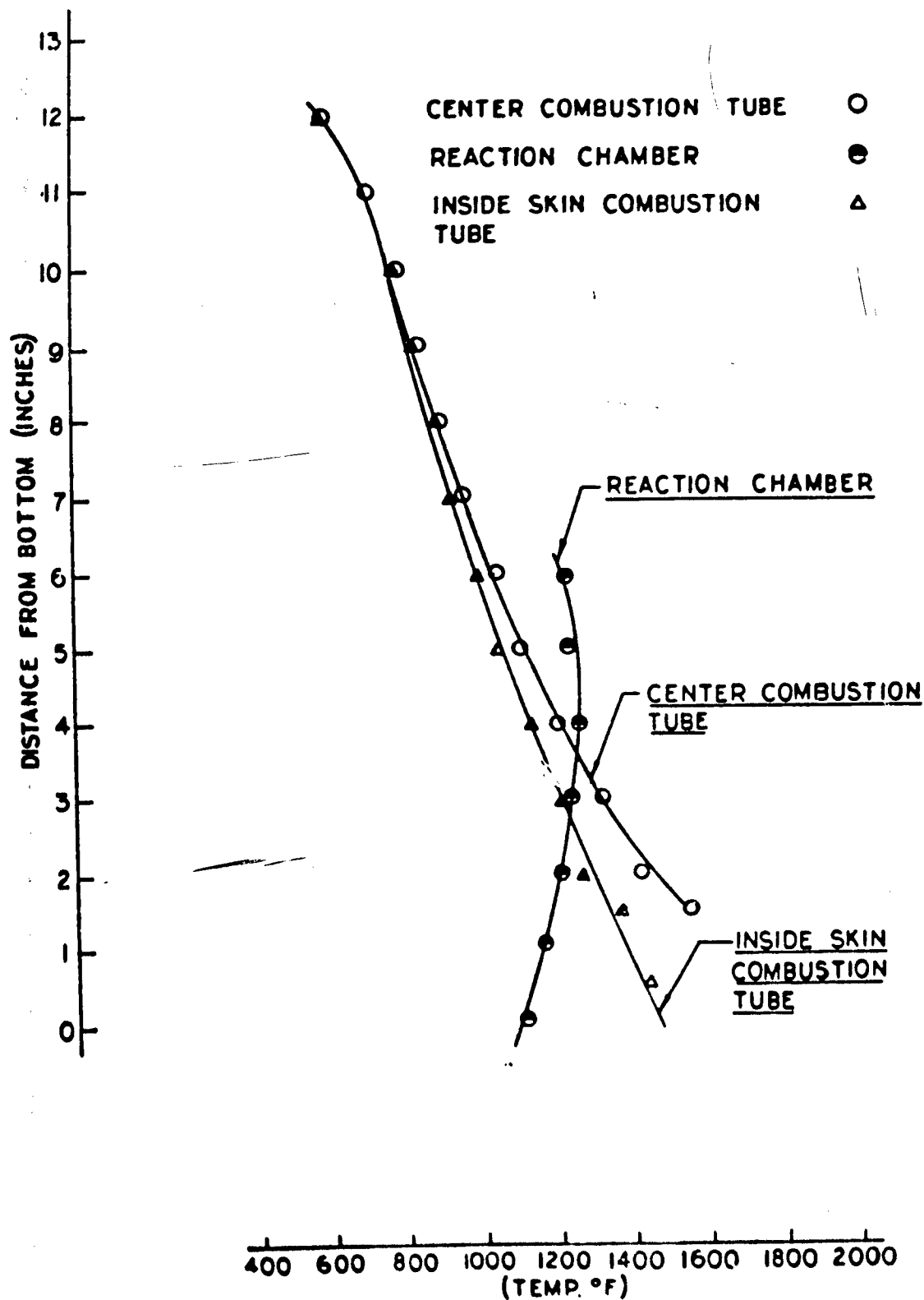


FIG. 9 **TEMPERATURE PROFILE** **PORTABLE H₂ GENERATOR**

PT.-5
COUNTERCURRENT FLOW

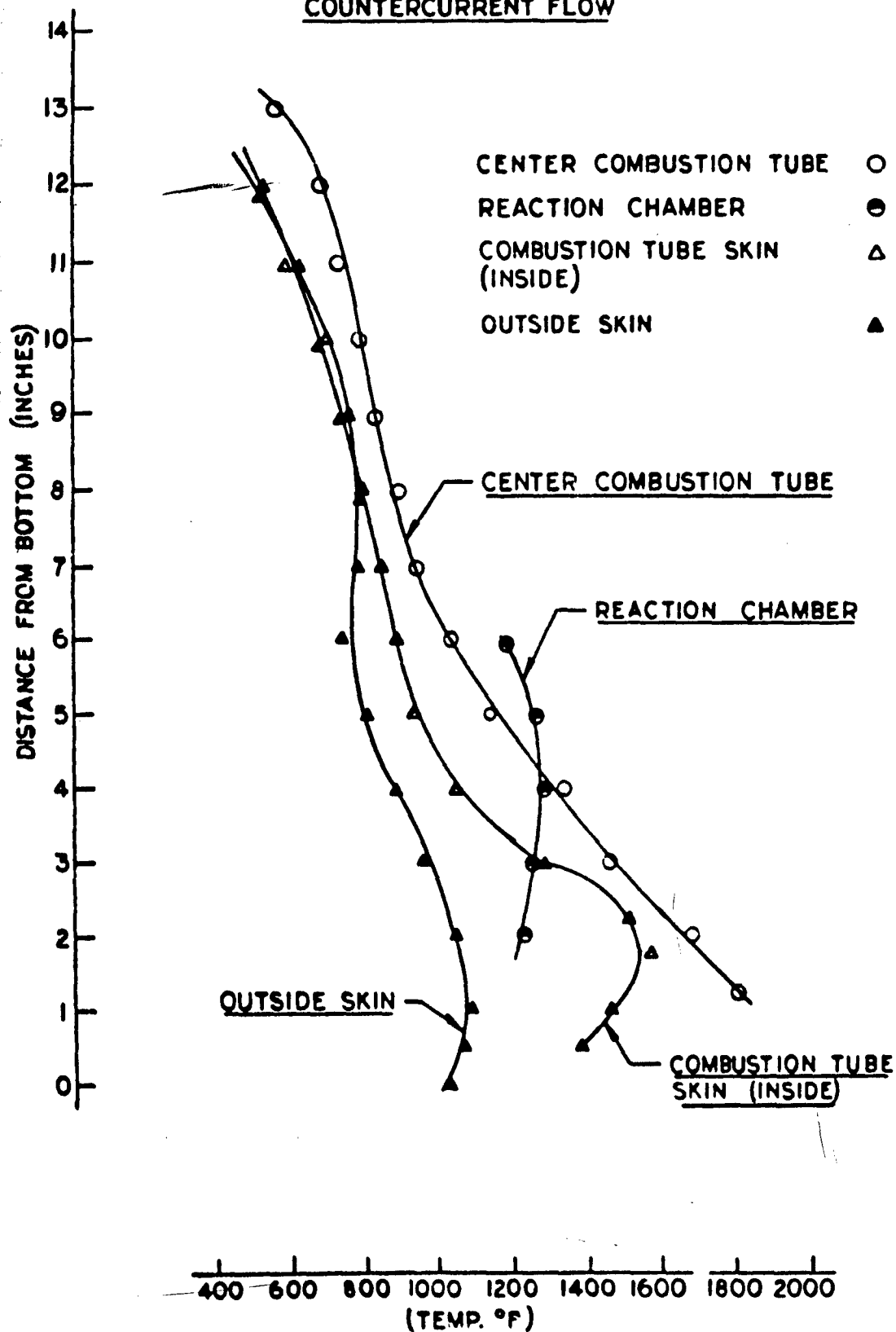


FIG. 10
SCHEMATIC FLOW DIAGRAM
TEST SET UP SPECIAL
EXPERIMENTAL PORTABLE H₂ GENERATOR
AMMONIA FUELED

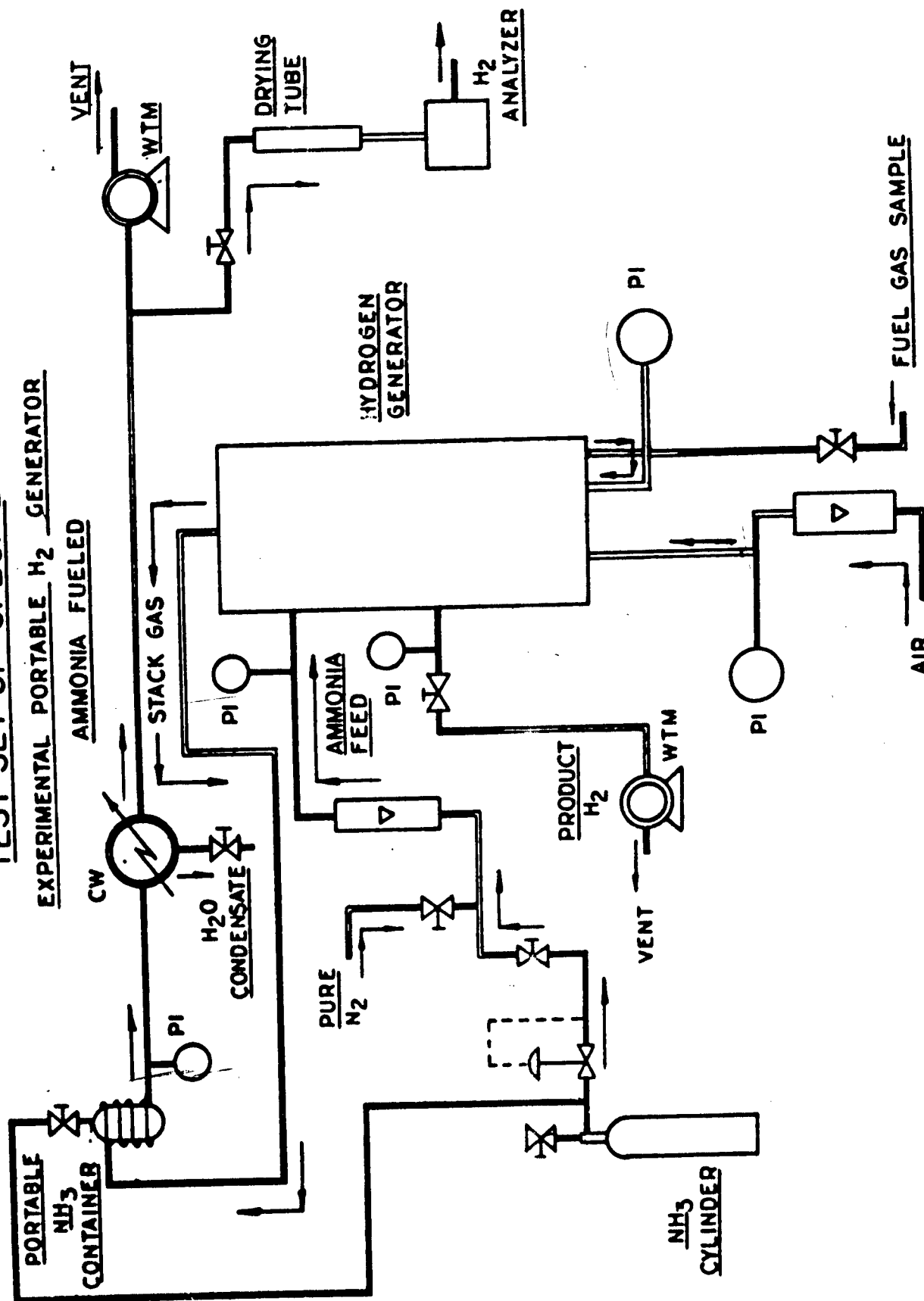


FIG. 11
AMMONIA CONTAINER
WITH
HEAT EXCHANGER JACKET

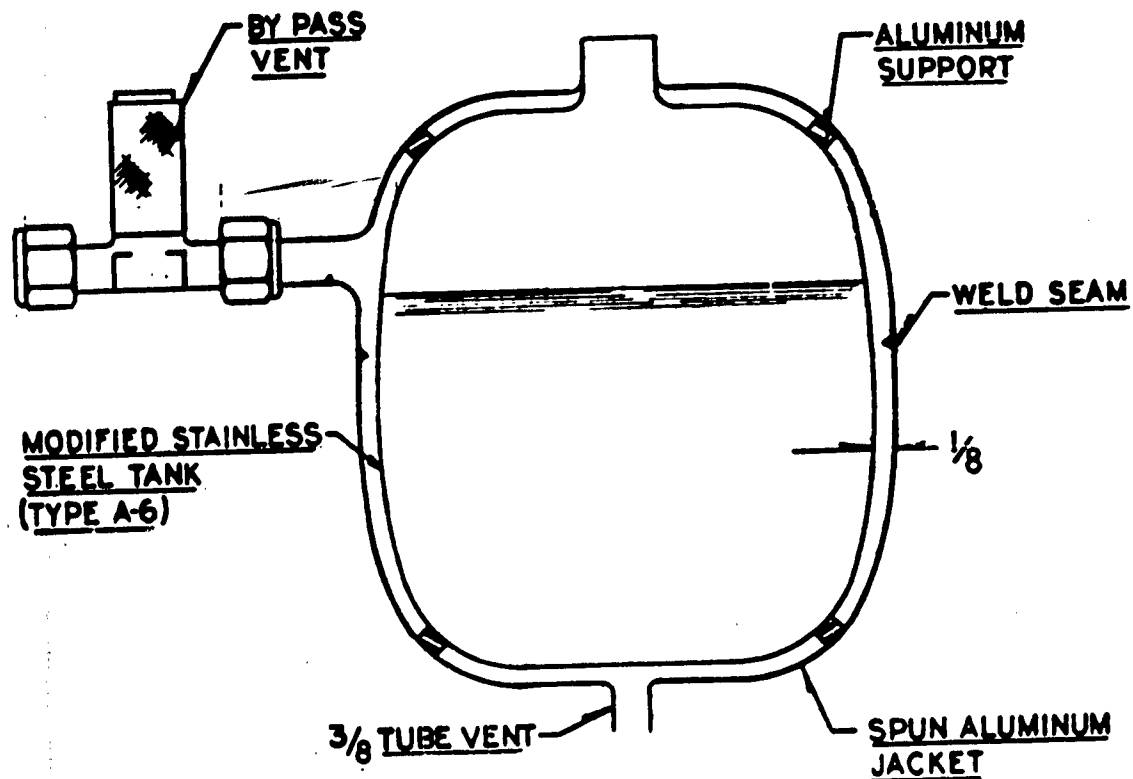


FIG. 12
ASSEMBLY CONFIGURATION
SAMPLE HYDROGEN GENERATOR DEVICE AMMONIA
FUELED

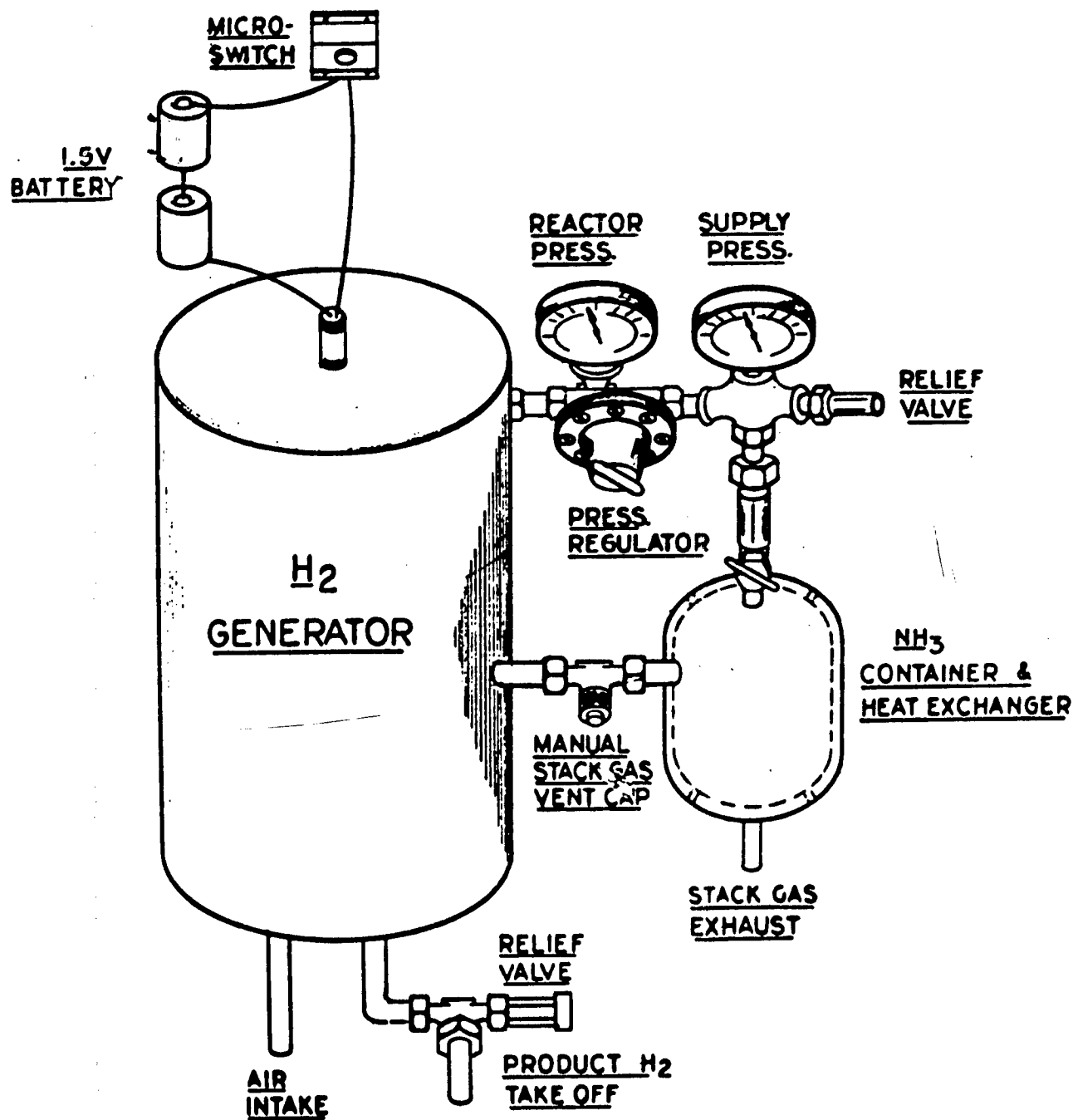


FIGURE 13

SCHEMATIC FLOW DIAGRAM

TEST SET UP

HYDROGEN GENERATOR DEVICE AMMONIA FUELED

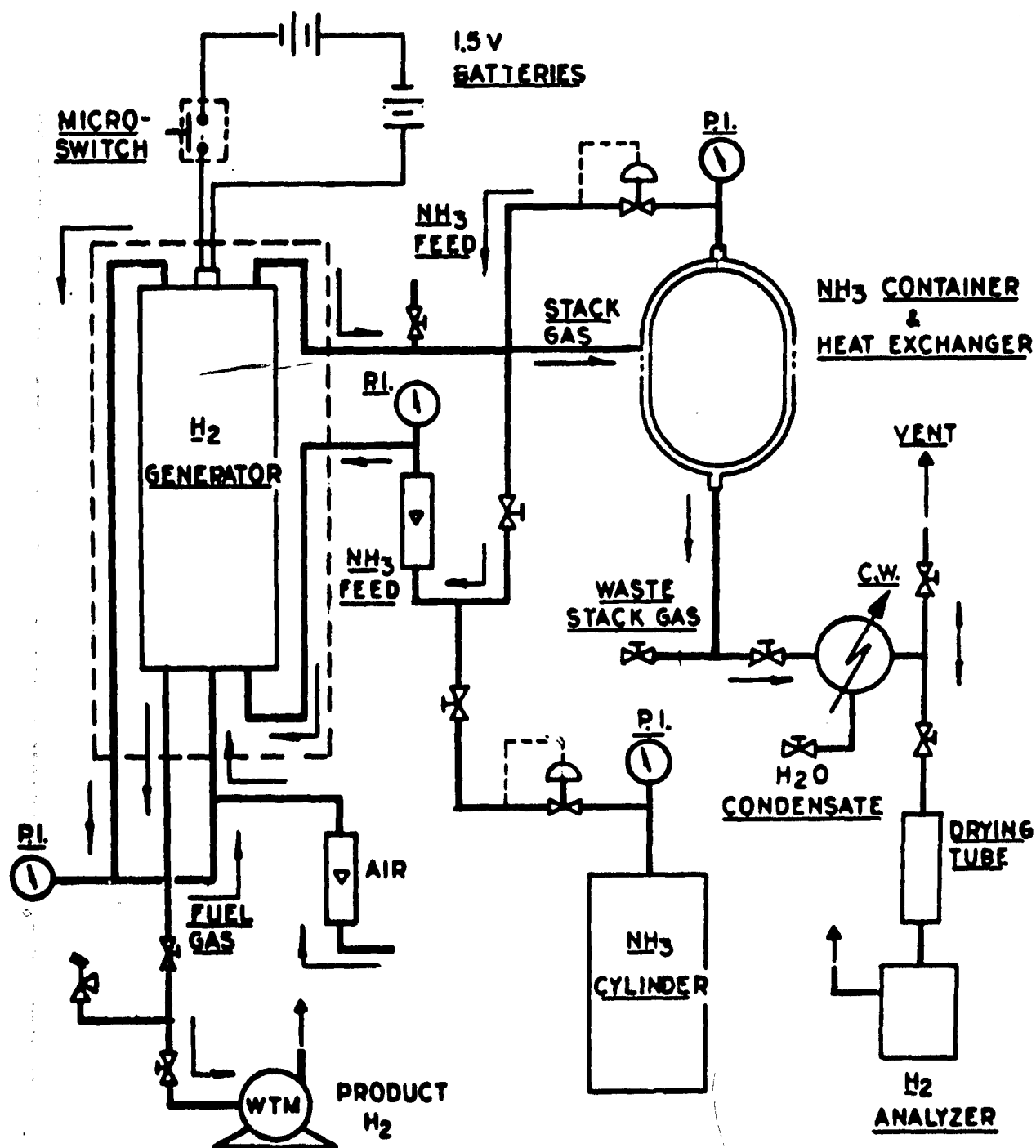


FIGURE 14

REACTOR TUBE PHOTOMICROGRAPH

Area of Failure

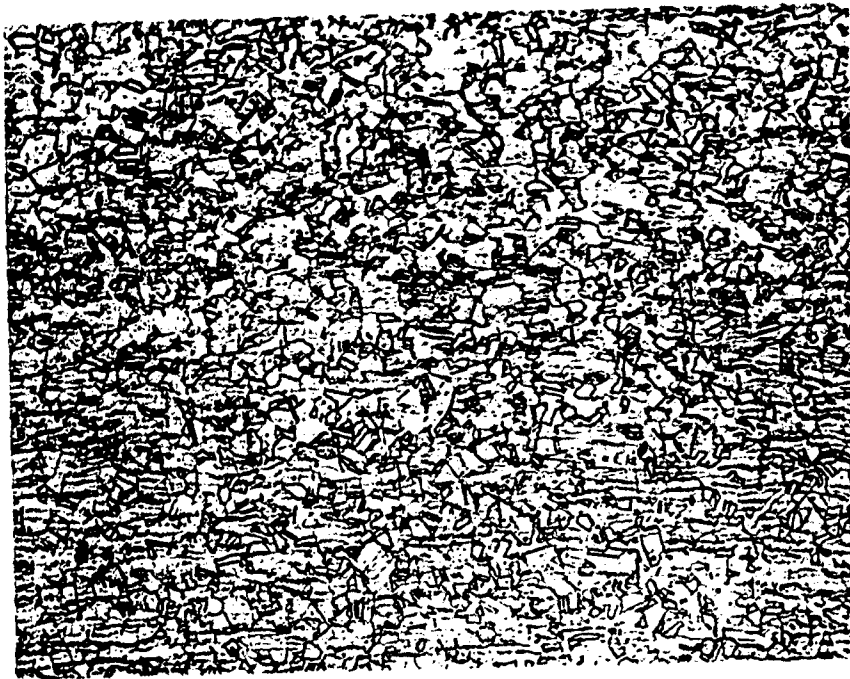


Magnification - 100 X

FIGURE 15

REACTOR TUBE PHOTOMICROGRAPH

Upper Portion of Tube



Magnification - 100 X

FIG. 16
SCHEMATIC FLOW DIAGRAM
TEST SET UP
METHANOL REFORMING

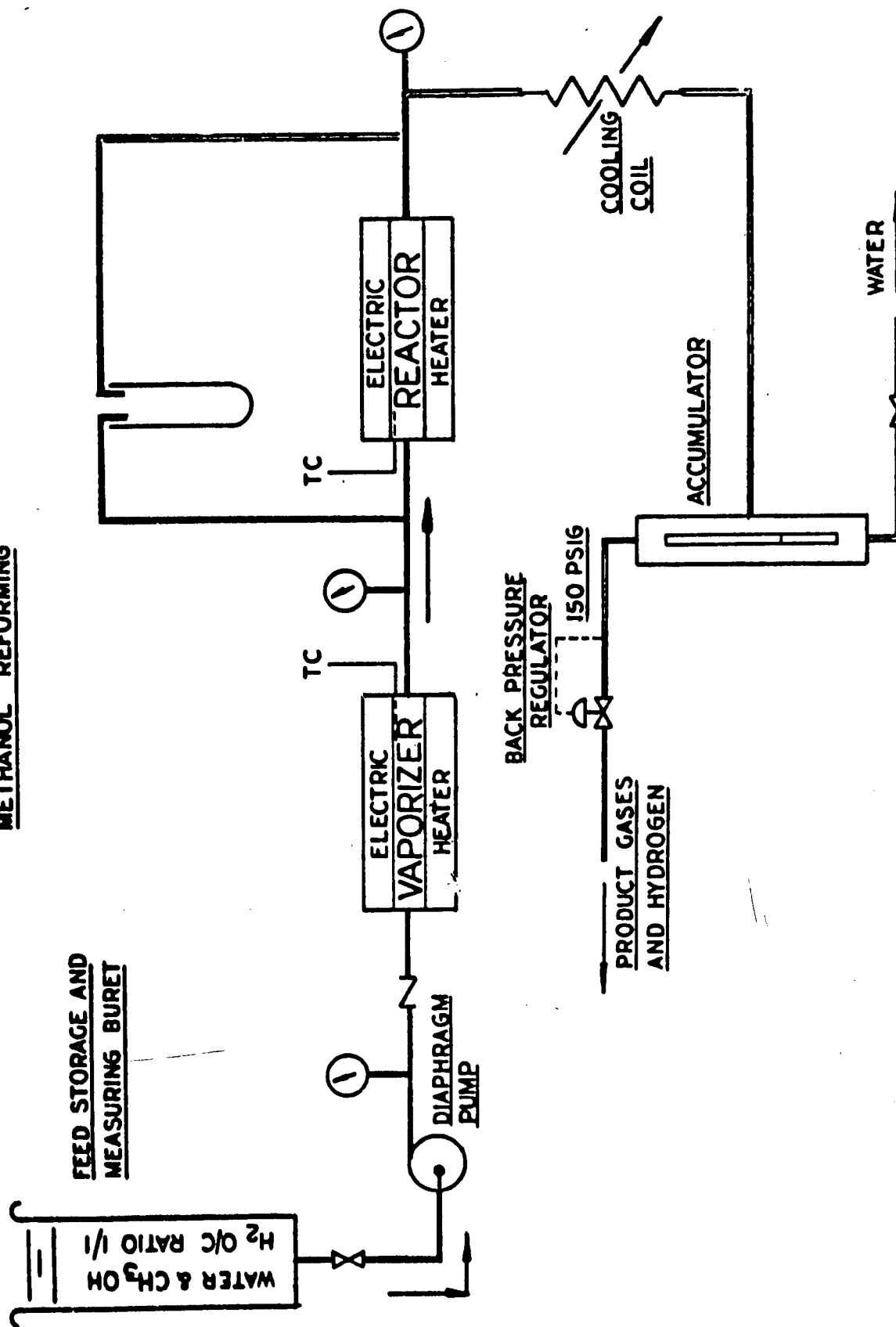


FIGURE 17
SCHEMATIC FLOW
DIAGRAM
TEST SET UP

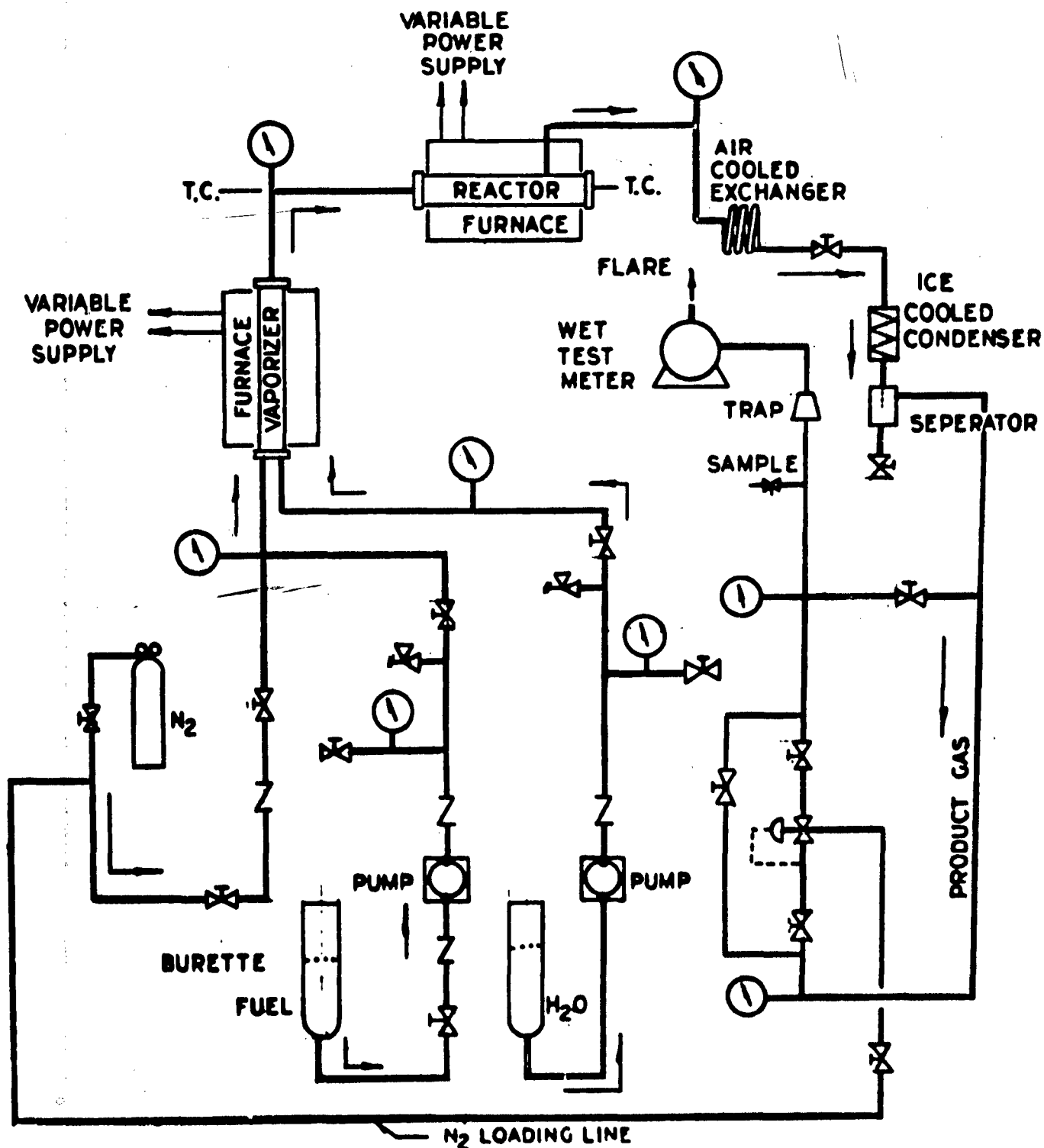
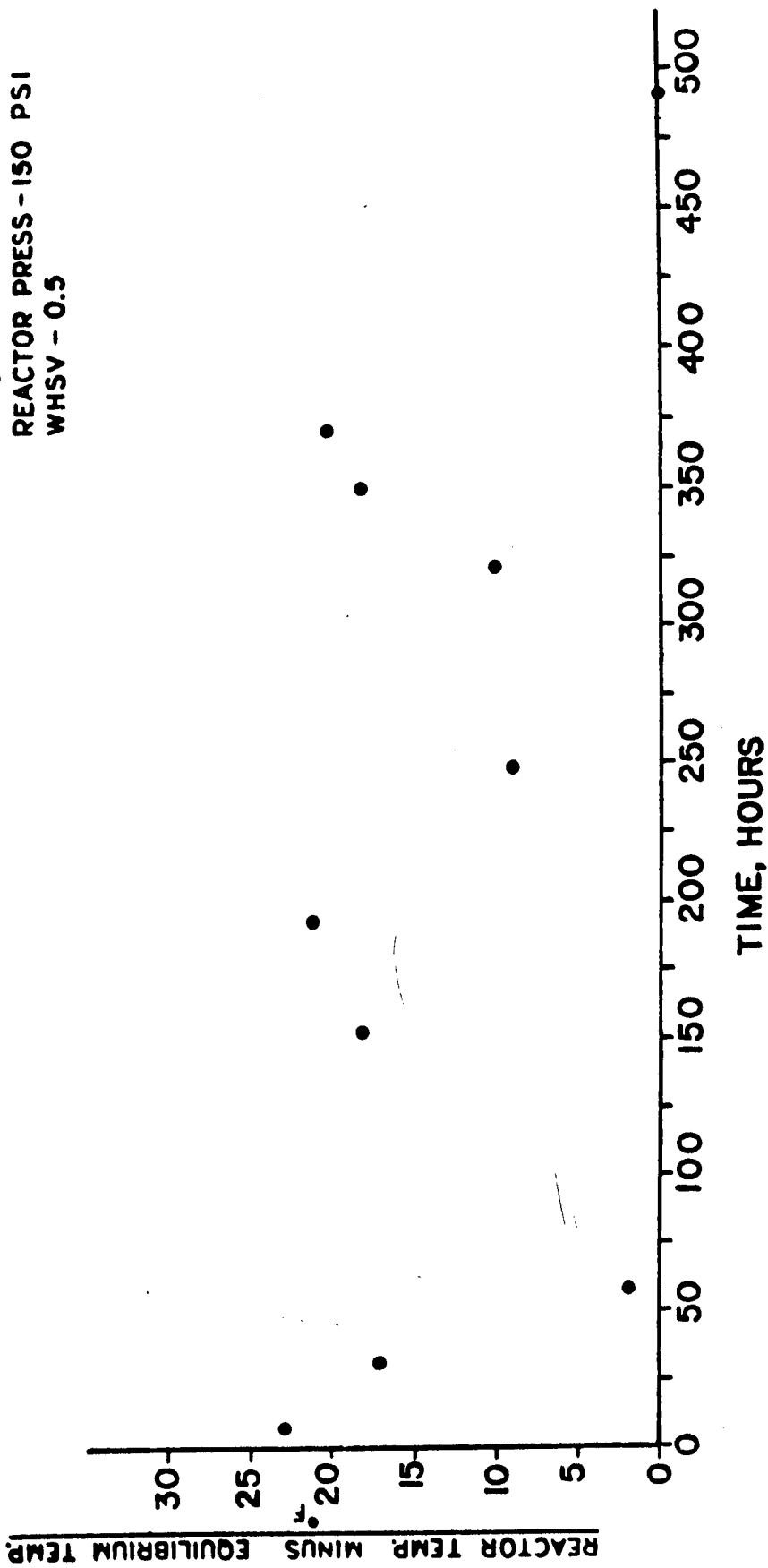


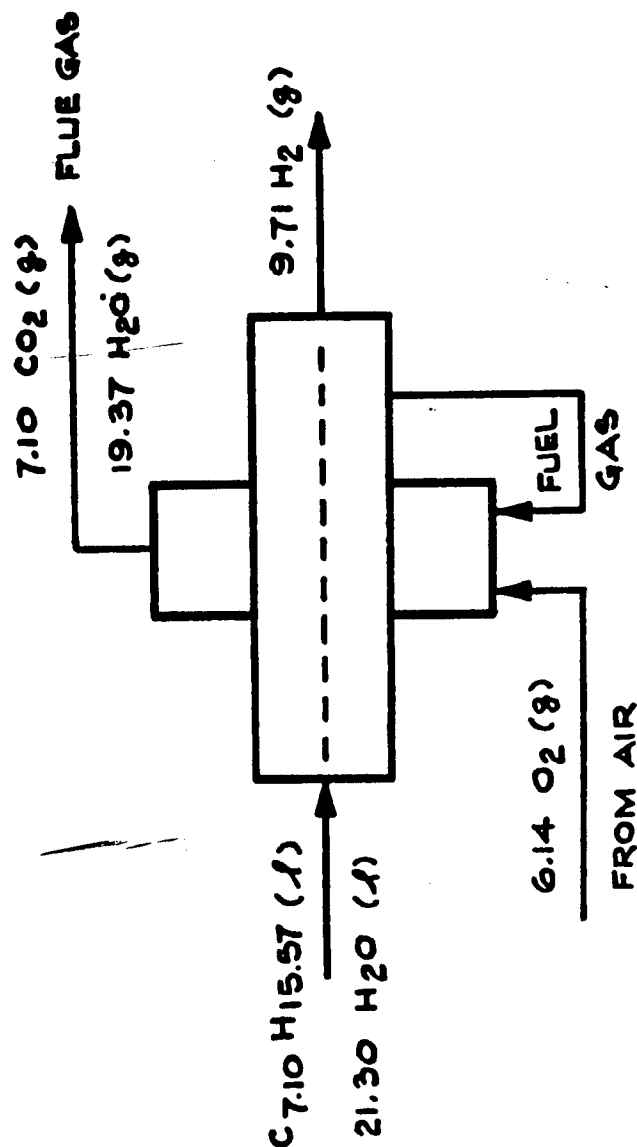
FIGURE 18

APPROACH TO EQUILIBRIUM

CONTINUOUS RUN
REACTOR TEMP - 1350 °F
REACTOR PRESS - 150 PSI
WHSV - 0.5



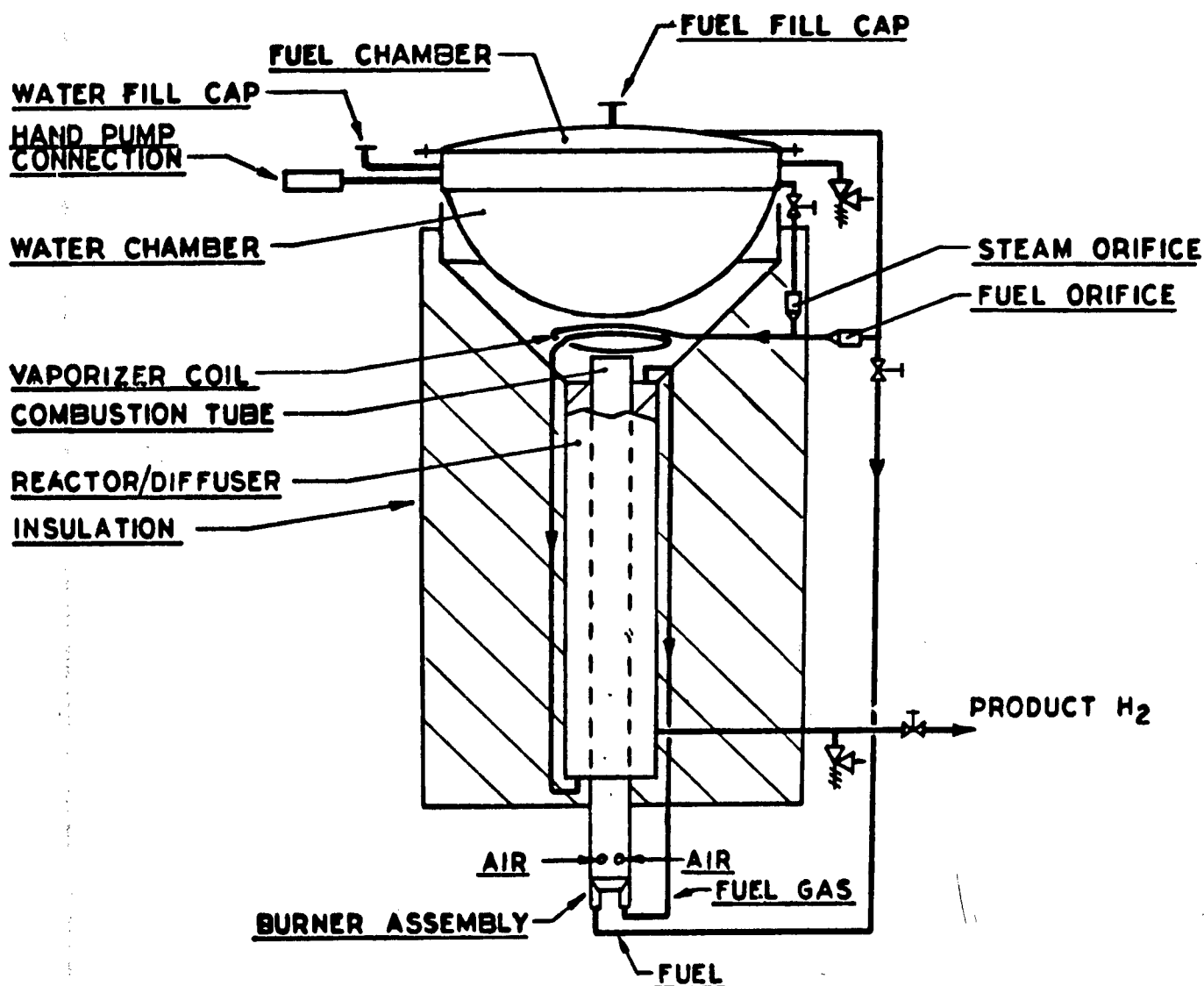
ENGELHARD HYDROGEN PROCESS



C_{7.10} H_{15.57}(l) + 21.30 H₂O(l) + 6.14 O₂(g) REACT OVER/ALL AND IN SEQUENCE TO FORM 7.10 CO₂(g) + 19.37 H₂O(g) + 9.71 H₂(g) PLUS A HEAT RELEASE OF 5,384 BTU/LB OF BTX INDEX RAFFINATE FUEL

FIGURE 19

FIGURE 20
HYDROGEN GENERATOR
LIQUID HYDROCARBON FUELED



HYDROGEN GENERATOR LIQUID HYDROCARBON FUELED 10 SCFH

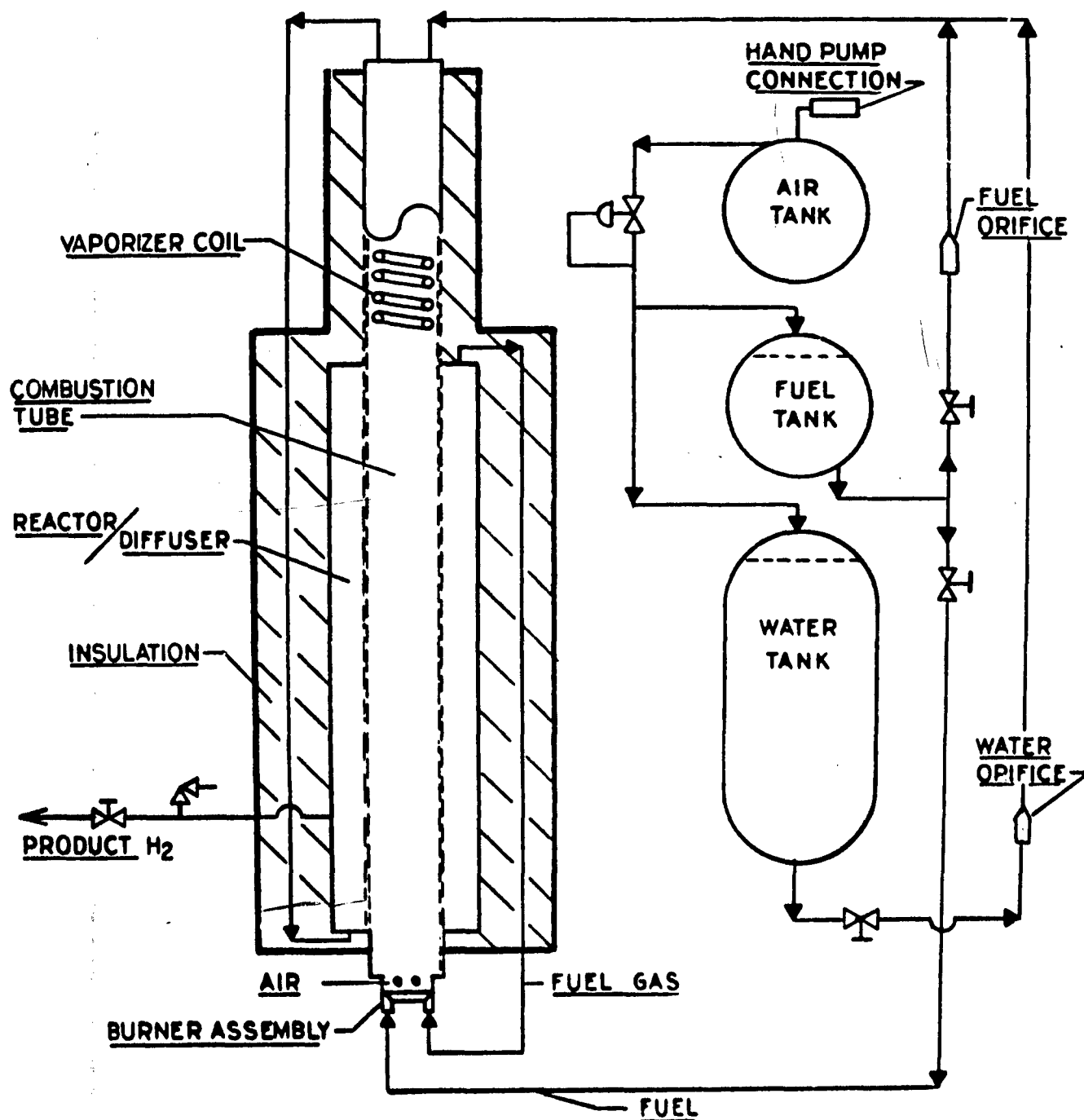


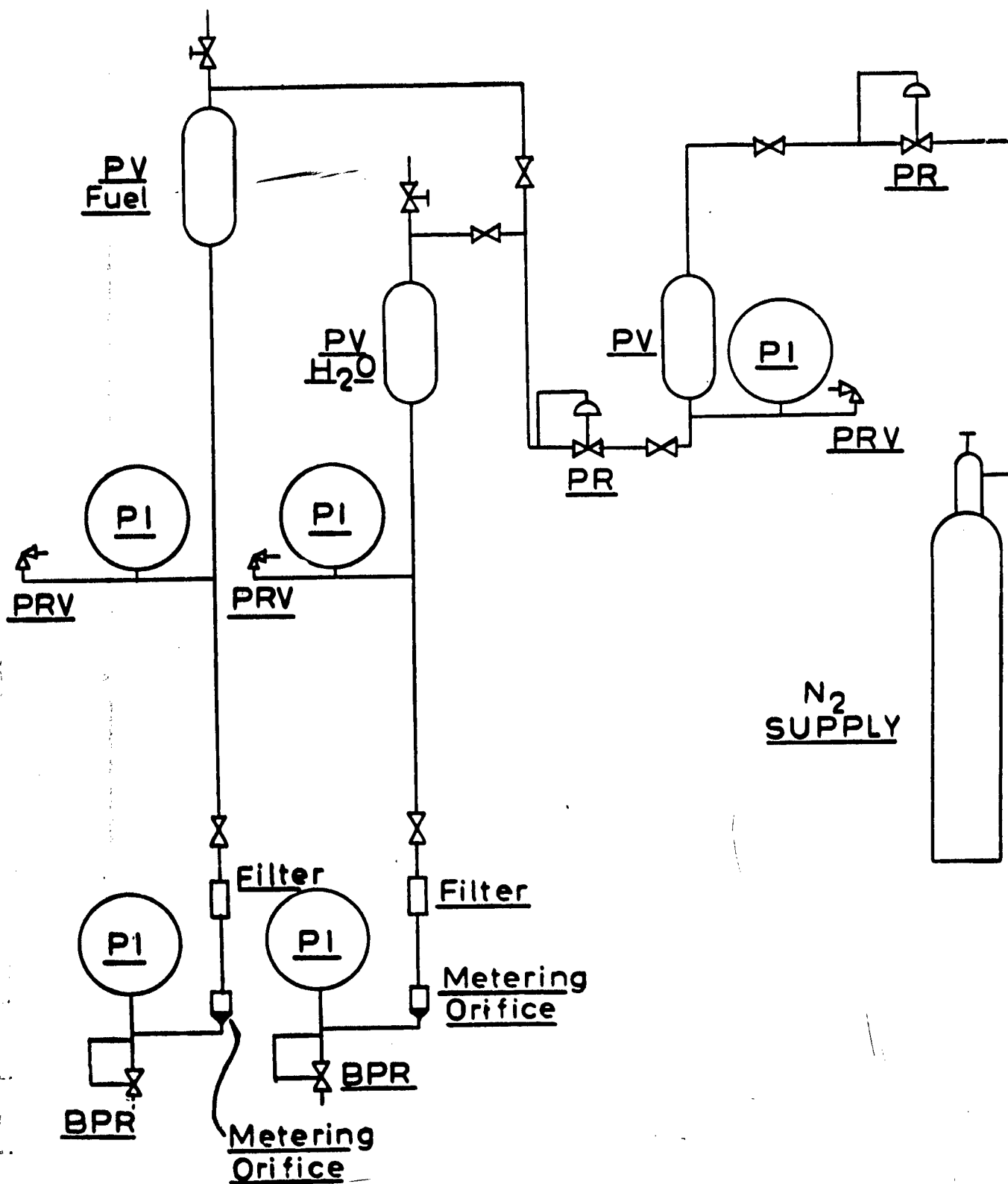
FIGURE 21

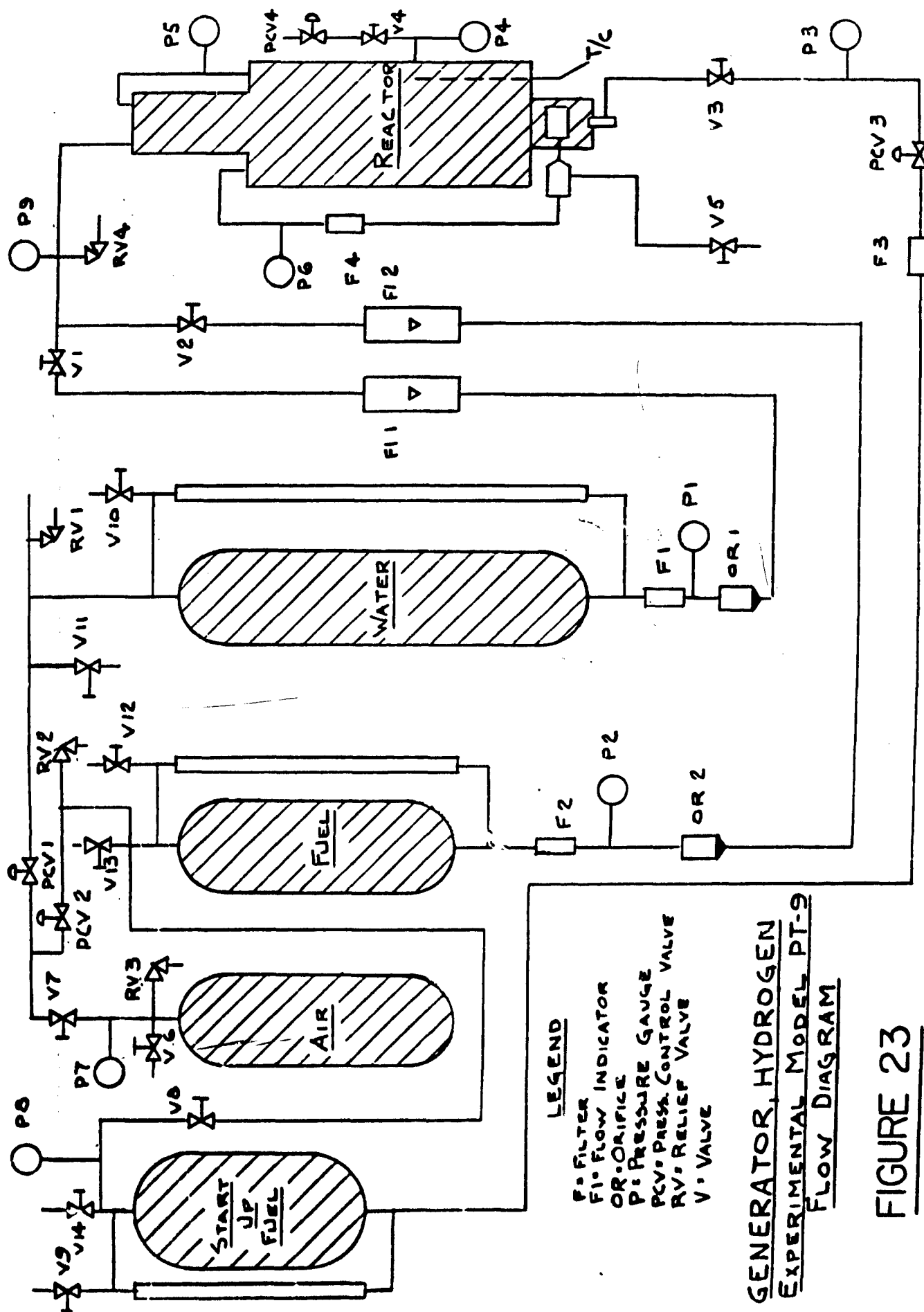
ENGELHARD INDUSTRIES, INC.

METERING ORIFICE TEST

FLOW DIAGRAM

FIGURE 22





GENERATOR, HYDROGEN
EXPERIMENTAL MODEL PT-9
FLOW DIAGRAM

FIGURE 23

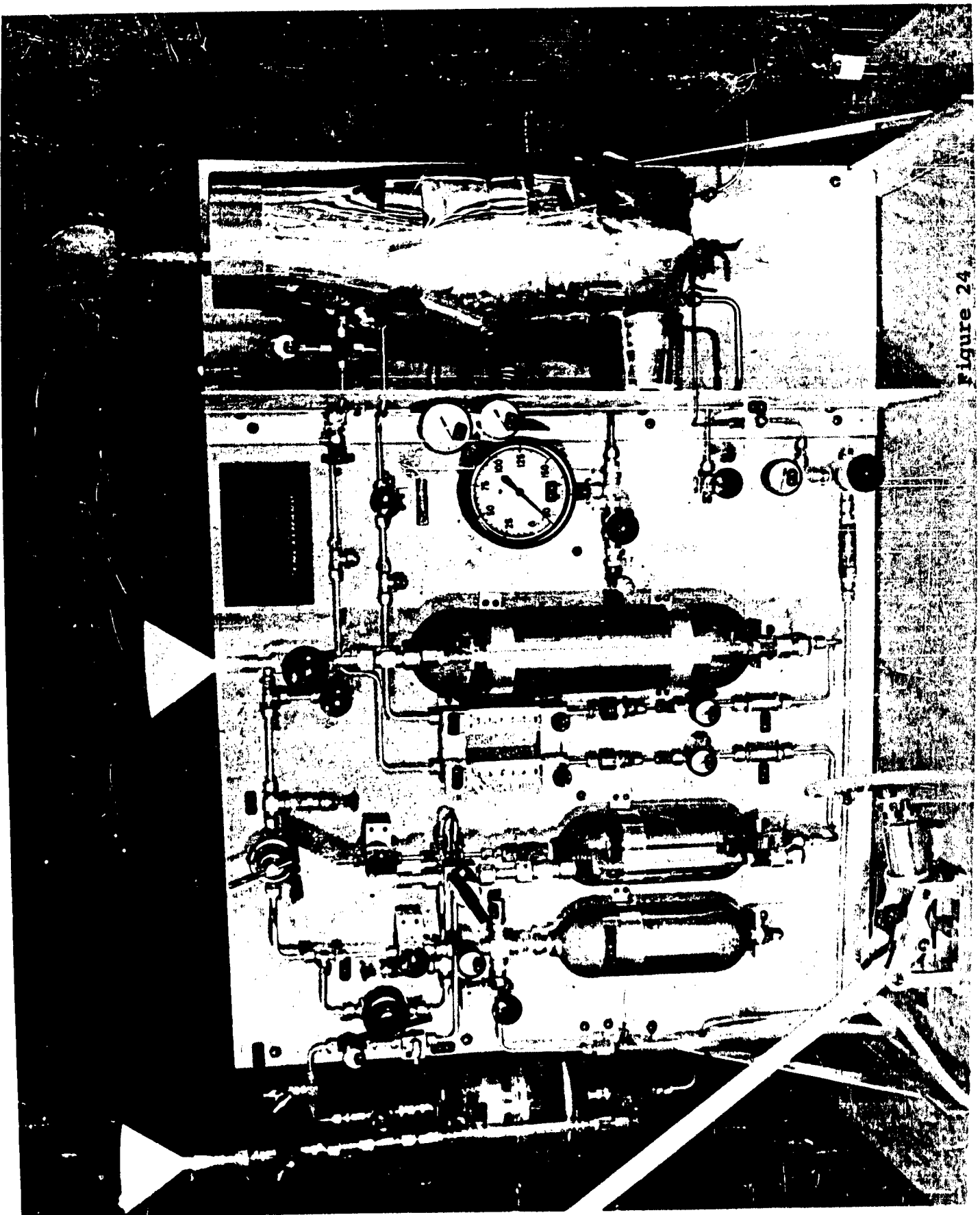


Figure 24

Engelhard Industries, Inc.
Process Equipment Division
Newark, New Jersey
HYDROGEN GENERATION FOR FUEL CELLS
by E. J. Emerson, L.E. Goodman,
H.H. Geissler, Rept. No. 8, Final Report
1 May 1962 - 30 September 1964
78 p. incl. illus. tables
(Contract DA-36-039 SC-89077)

UNCLASSIFIED REPORT

A research project is carried out to develop a method of generating hydrogen from economical fuels (including ammonia, methanol, and liquid hydrocarbons) in such a manner that the generation of hydrogen and its separation from other reaction products can be accomplished in the field on a portable basis. The Engelhard Hydrogen Process is used which combines a hydrogen producing reaction, removal of the pure product hydrogen, and use of residue as fuel to supply the energy requirements of the process. Life tests of 490 and 1035 hours were performed with ammonia fueled generators. A final prototype portable ammonia fueled generator, having a thermal efficiency of over 70%, was delivered to the U.S. Army Electronics Laboratory. Investigations have shown it is possible to steam reform methanol and liquid hydro-

(over)

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(over)

UNCLASSIFIED

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2. Hydrogen fuel preparation.
3. Portable fuel generator.

I. Emerson, E.J.
Goodman, L.E.
Geissler, H.H.
U.S. Army Electronics Laboratory.
III Contract DA-36-039 SC-89077

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